The Abdus Salam International Centre for Theoretical Physics

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CIMPA/ICTP Geometric Structures and Theory of Control

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Warm dense matter physics

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Thanks to
Richard M. More (LBNL)
Hikaru Kitamura (Kyoto U.)

I. Introduction

II. Detailed physical model for warm dense matter

III. New trends for warm dense matter research works and their applications

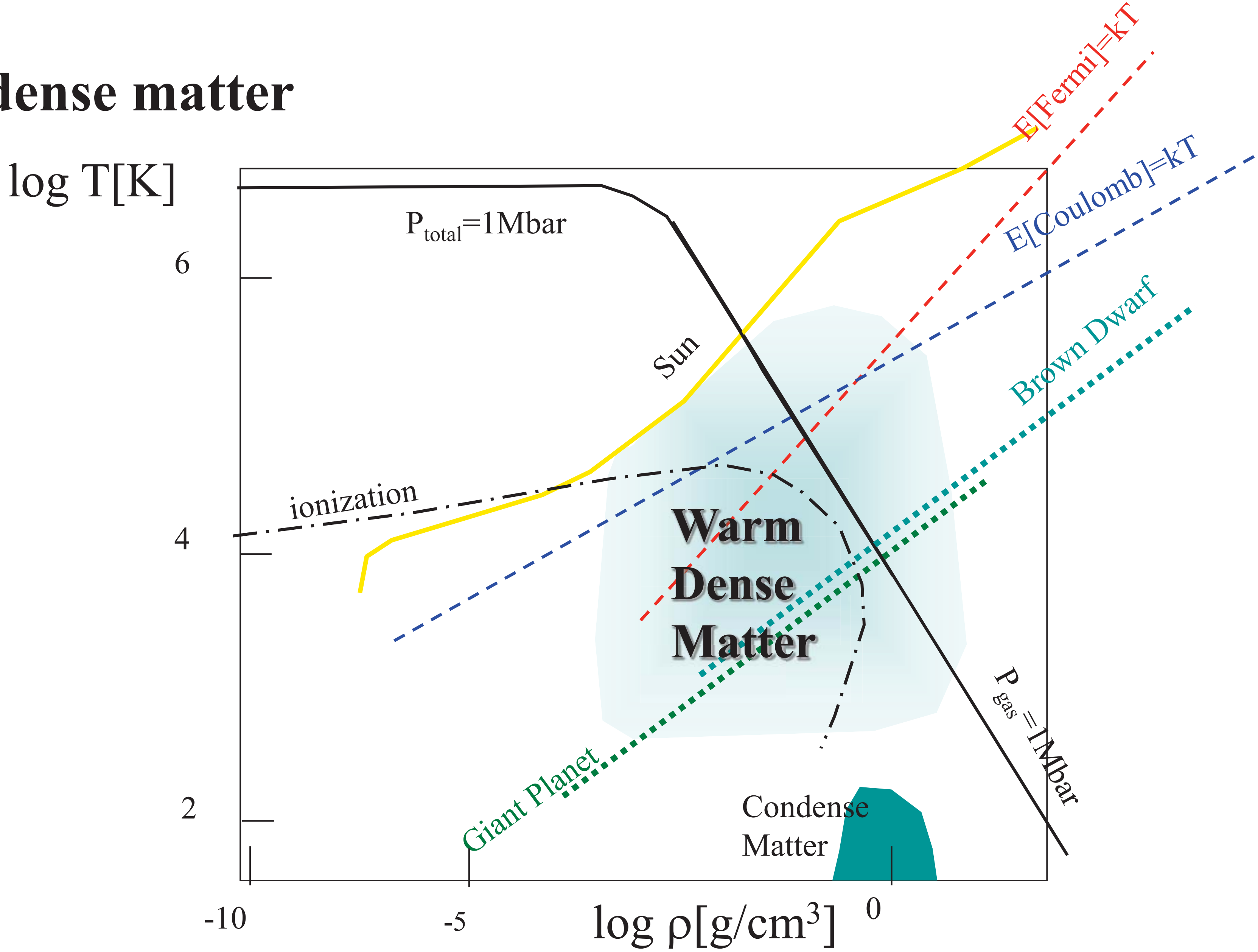
What subjects can we cover with this lecture?

- What is warm dense matter?
- History
- Extrapolation from known physic idea and its failing
- What should we do for understanding wdm?
  - Idea from condensed matter physics
  - Idea from plasma physics
- Understanding of modeling of wdm
- Recent progress in wdm research

What is WDM?

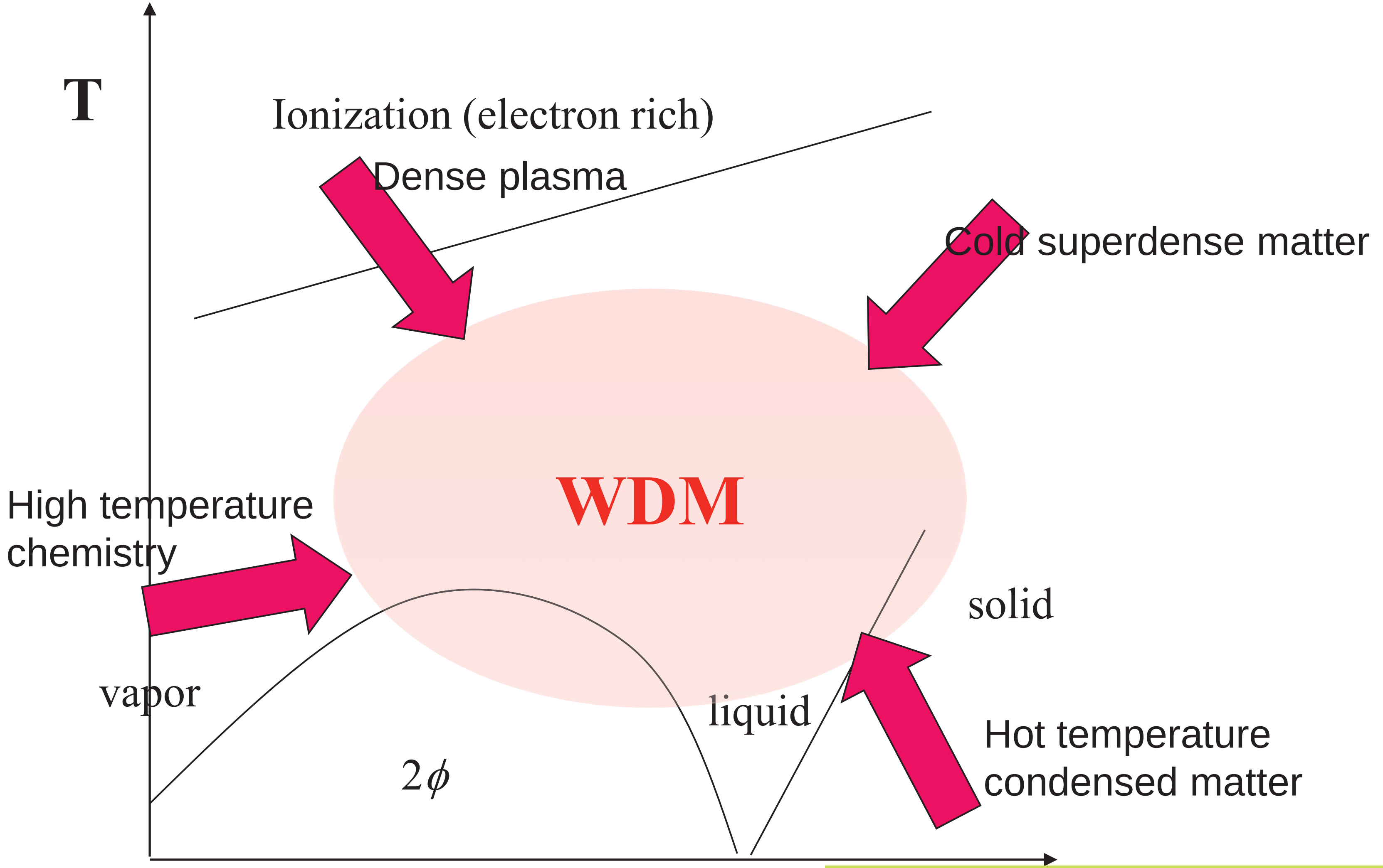
(brief definition)

Warm dense matter



*Chemical force(condensed matter) ~ Coulomb force(ideal plasma)
*Electron degenerated plasma (Giant planet interior material)
*Strongly coupled plasma
*Metal-insulator transition(minimum conductivity, Anderson-type transition?)
*Two phase region [gas and liquid] (droplet or debris formation)

Warm dense matter

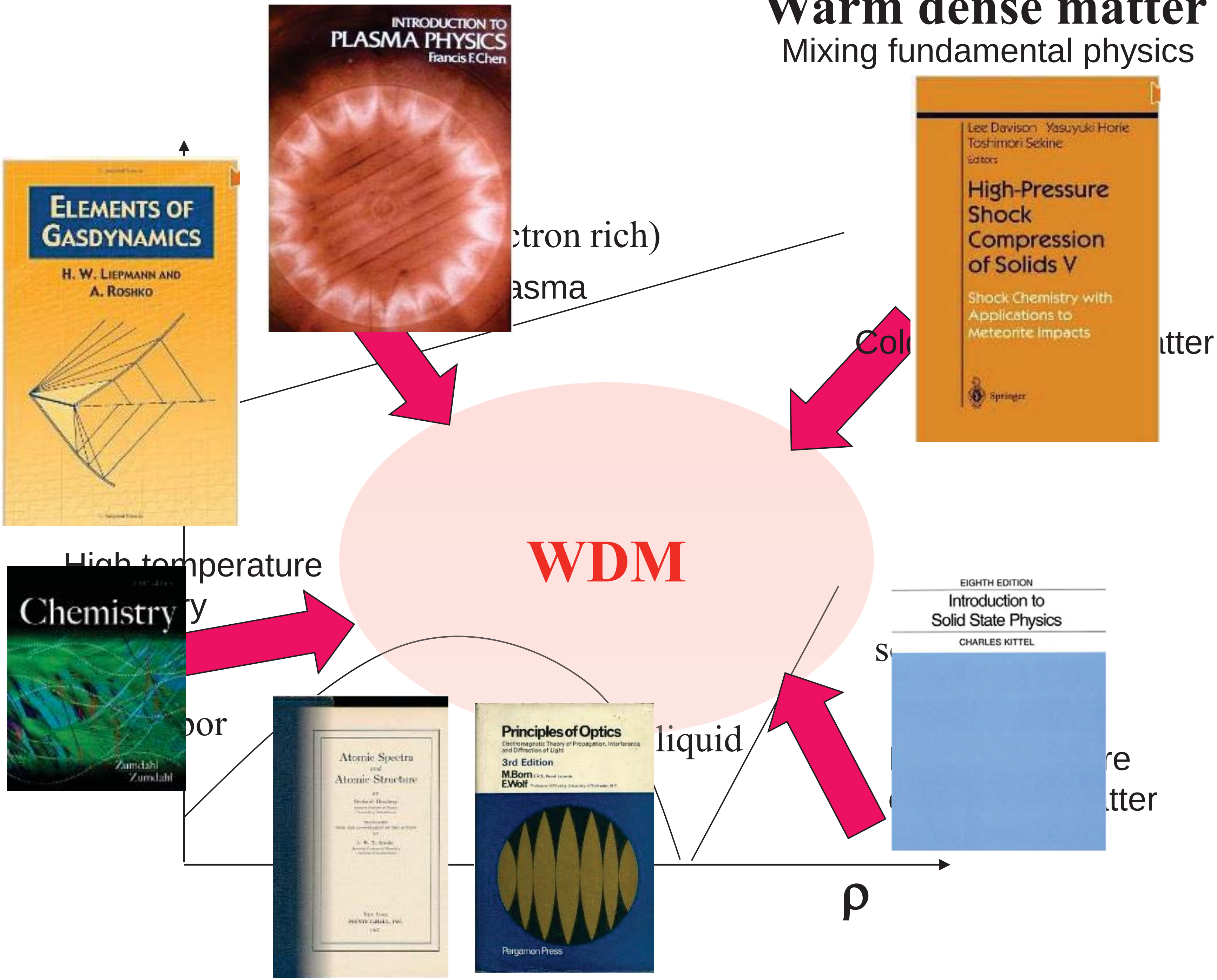


Strong inter atomic interaction + electronic excitations

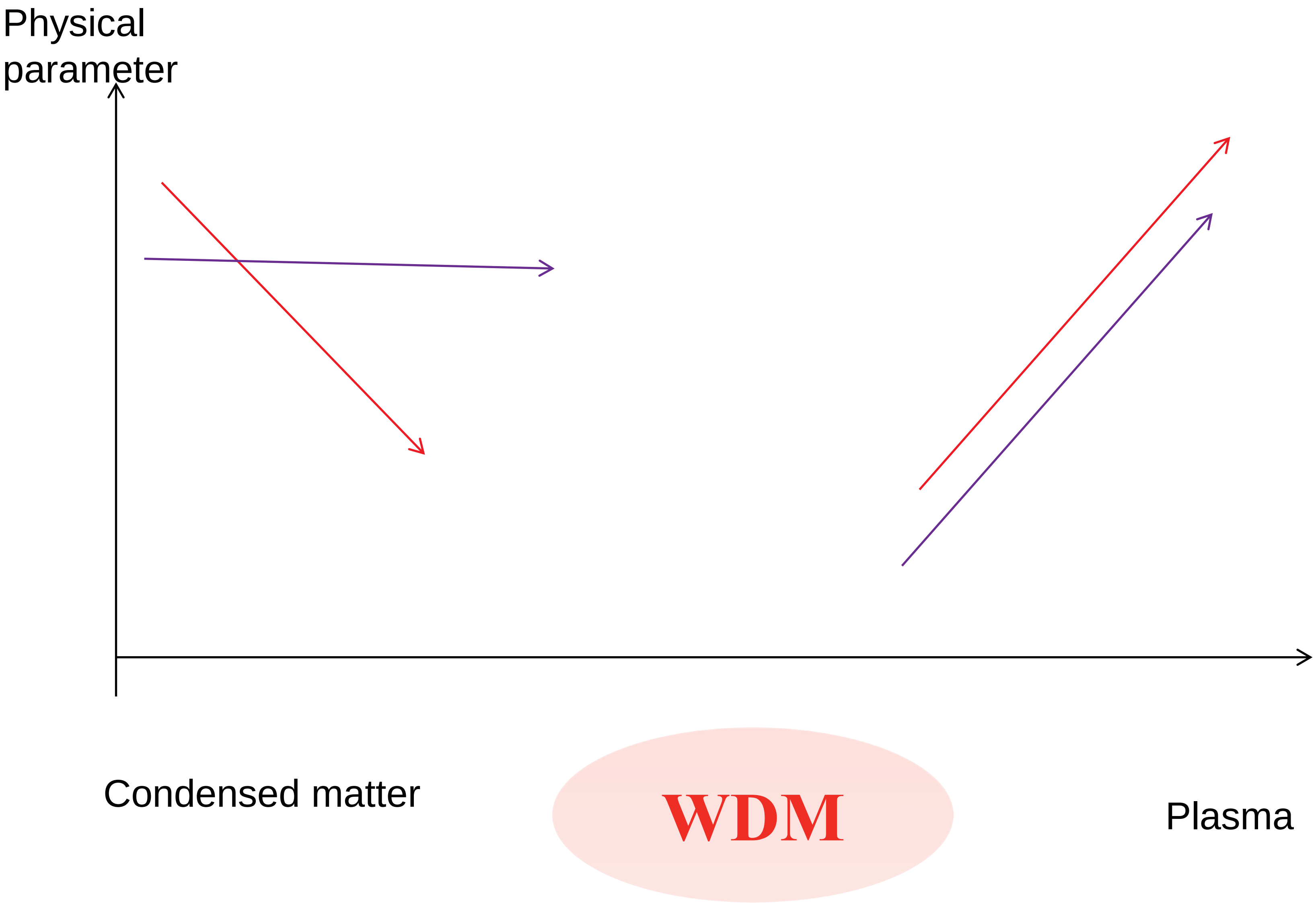
There are a lot of new physics and many uncertain phenomena.

Warm dense matter

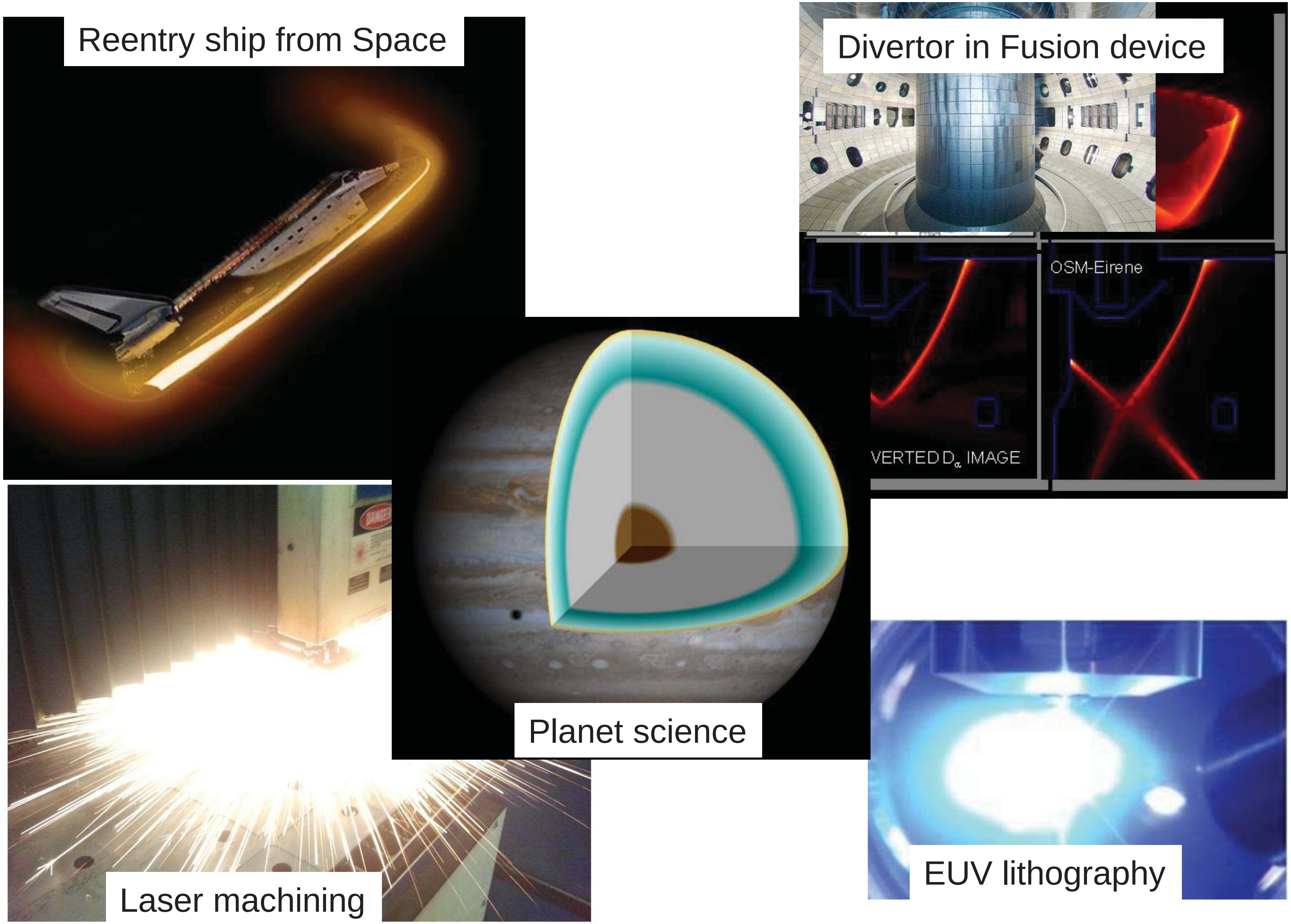
Mixing fundamental physics



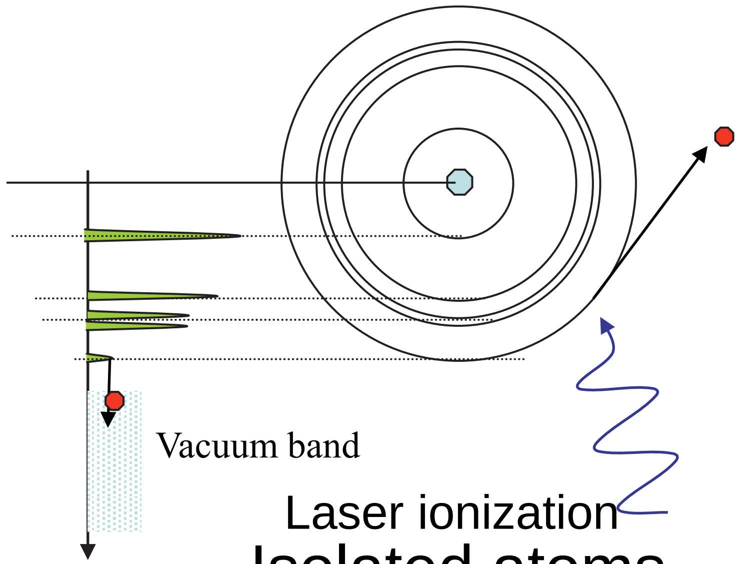
Some parameters in WDM are very different in Solids and Plasmas



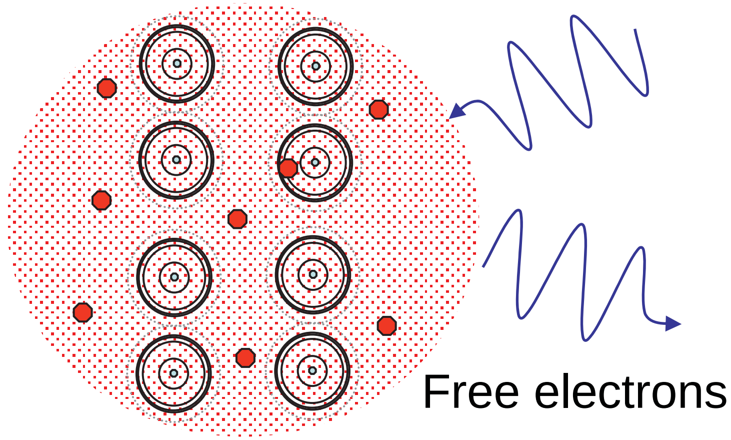
Understanding of Warm dense matter is now needed in many plane



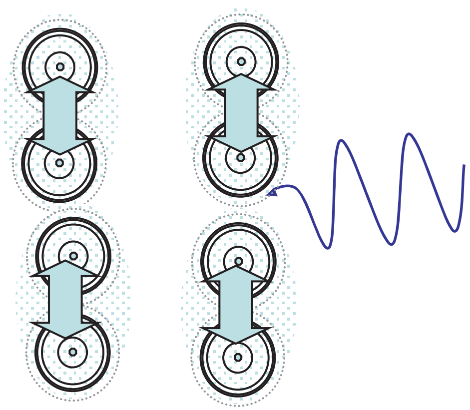
What is interesting in physics at this subject.



Isolated atoms

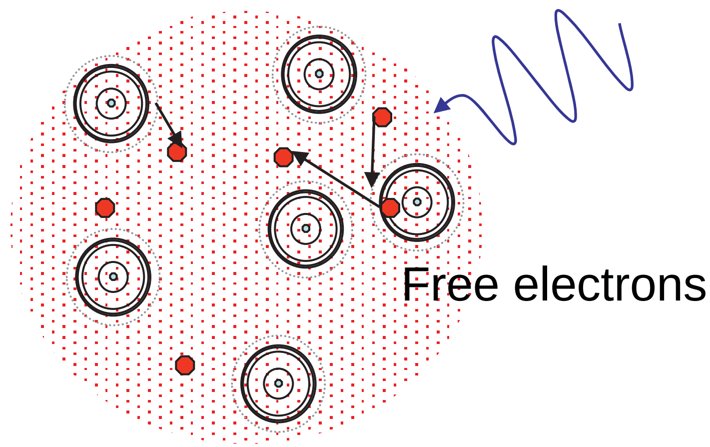


Metals



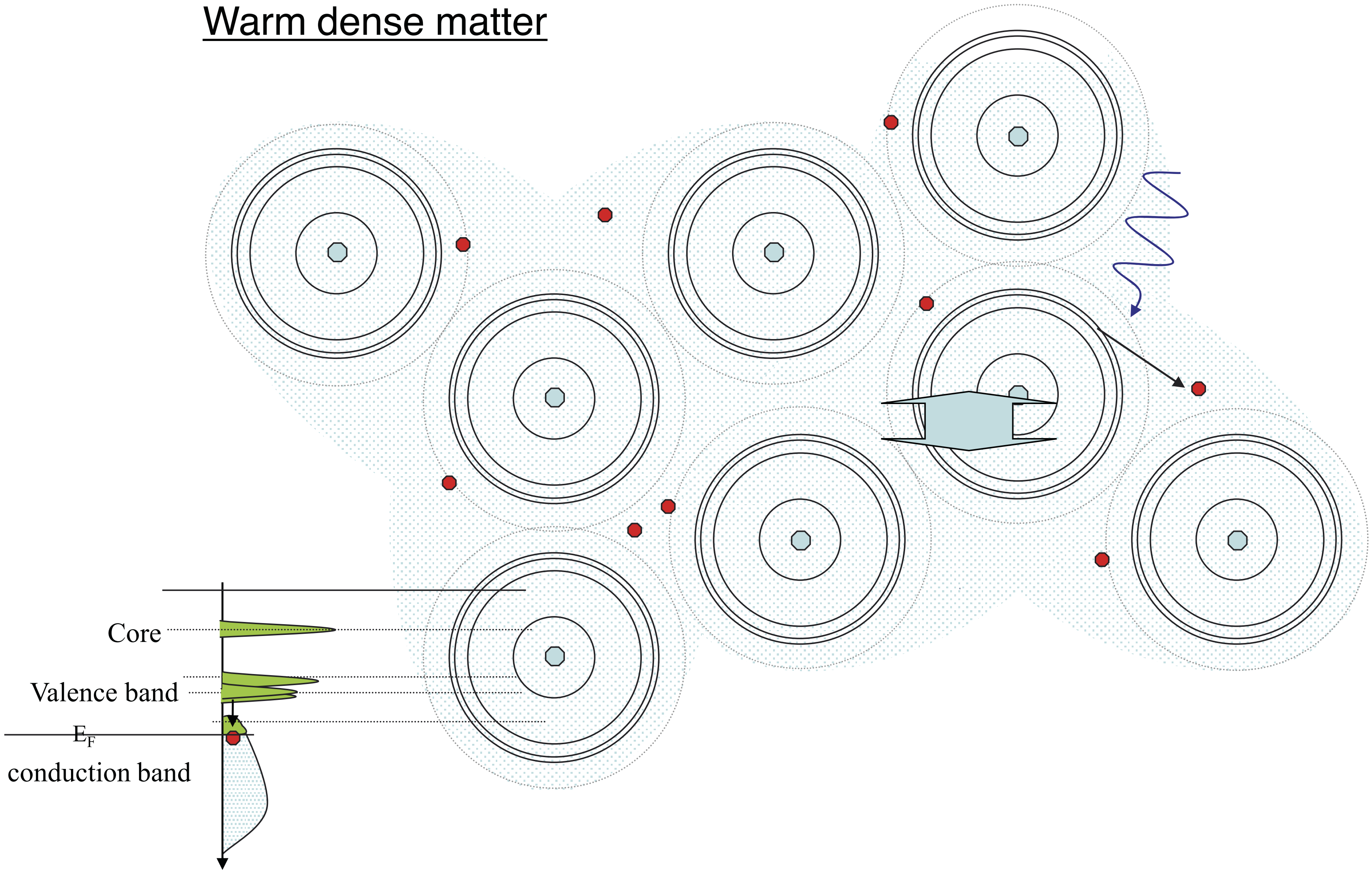
Laser induced polarization

Dielectric materials



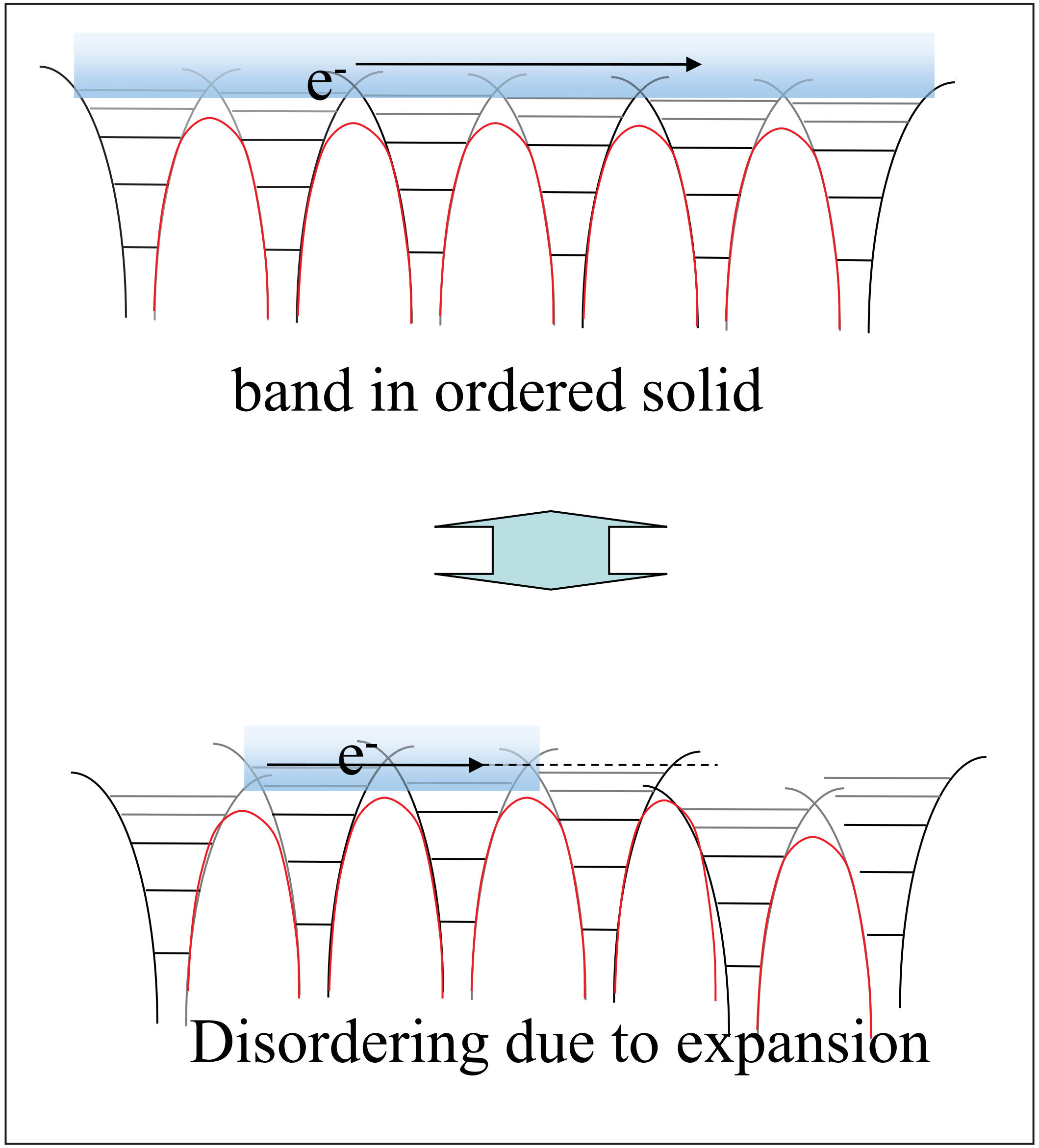
Ordinary plasmas

Warm dense matter



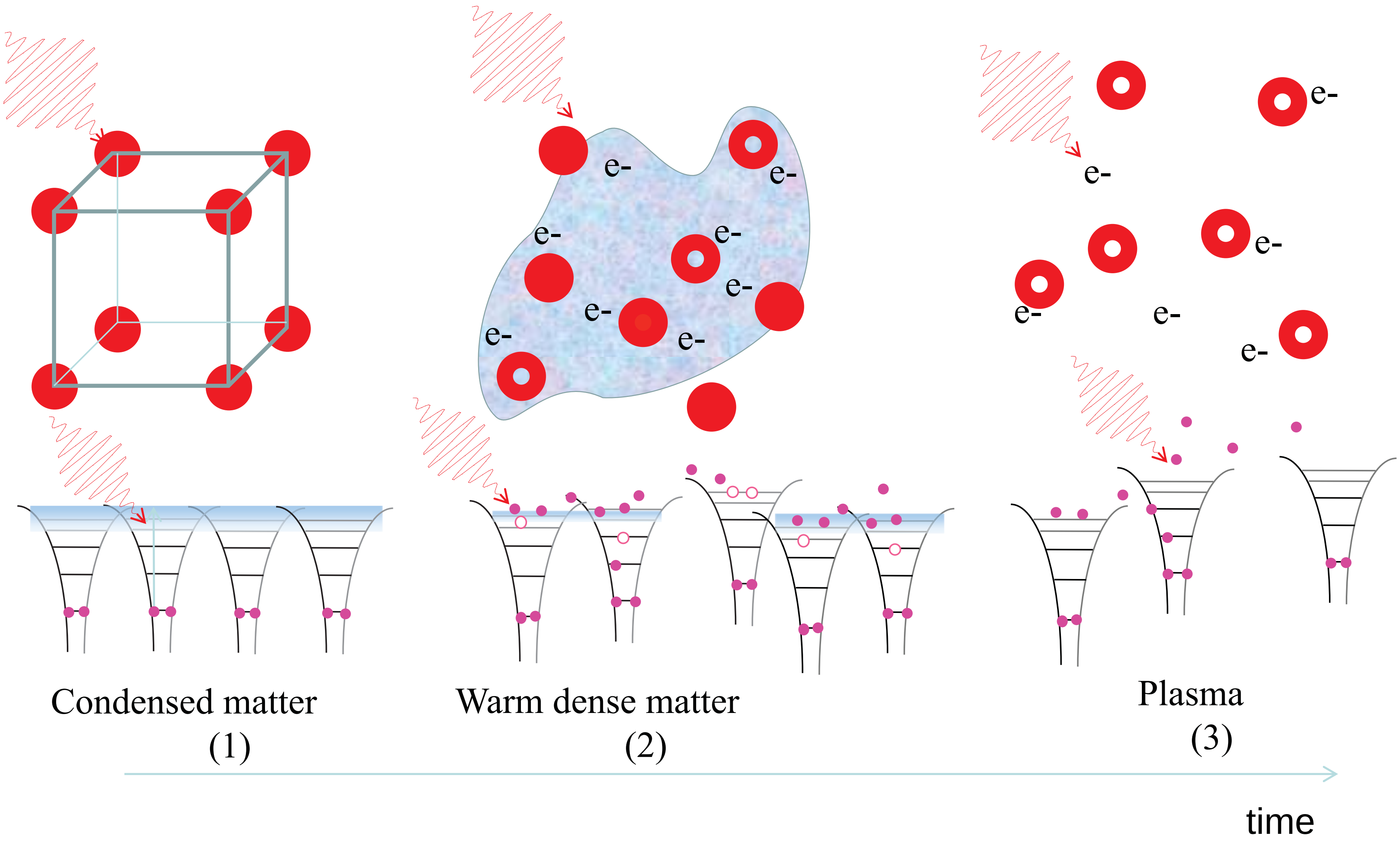
Ionization, Electron excitation but keeping lattice order

Key points: Deviation from free electron model



e-
band in ordered solid
e-
Disordering due to expansion

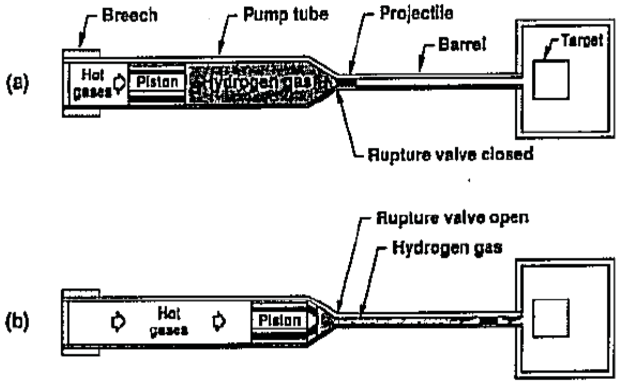
Material change after illumination of pulse lasers



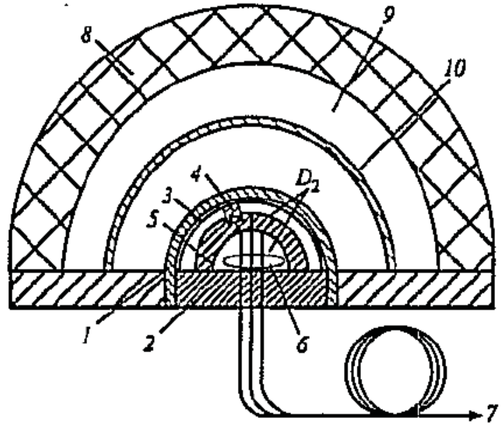
History

1st WDM experiments

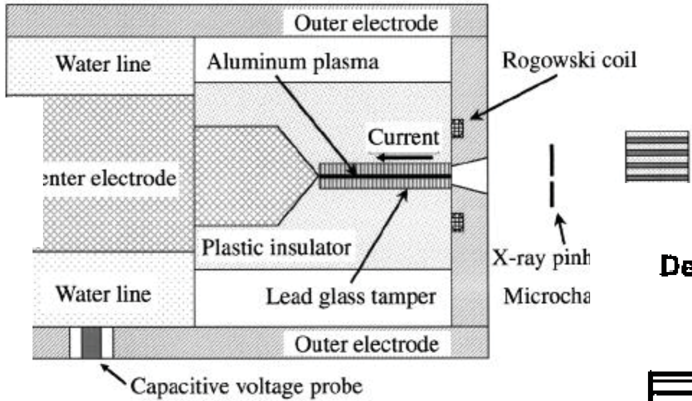
Gas gun



Converging shock

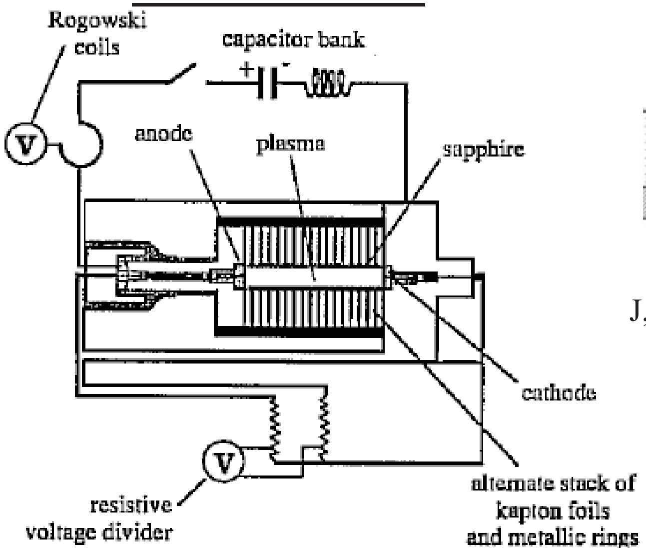


Fast discharge

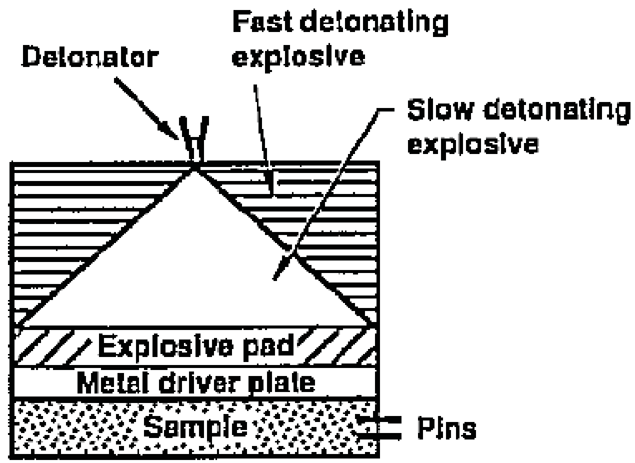


J,F. Benarge, et. Al., PRL 83-15 (1999)

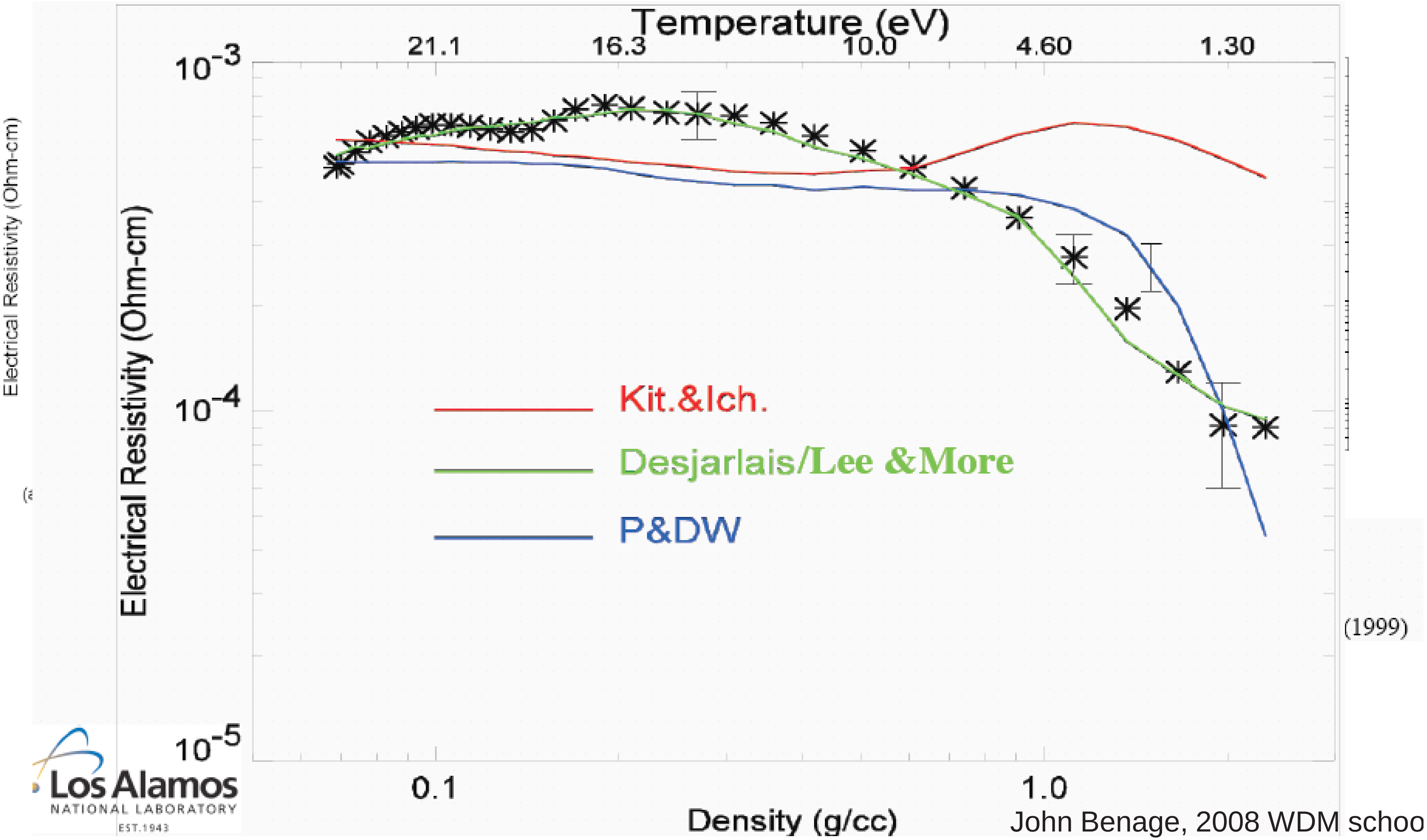
Plasma cell



explosive shock



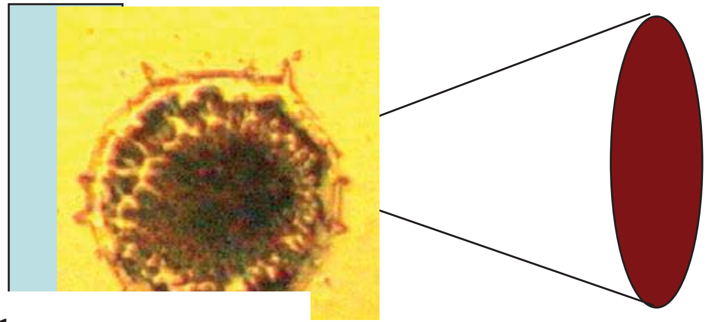
Accurate and reliable DC conductivity data!



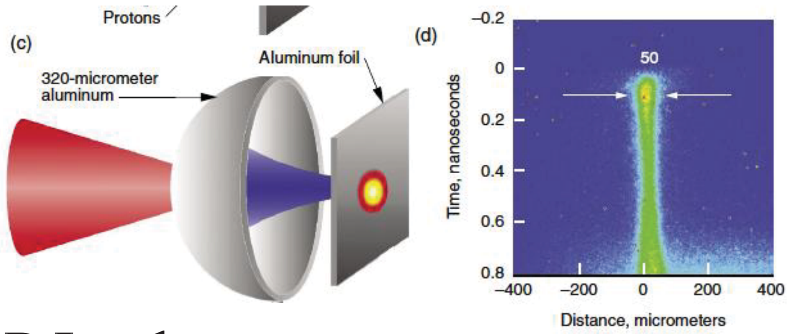
Los Alamos NATIONAL LABORATORY EST.1943

John Benage, 2008 WDM schoo

Variety of WDM experiments

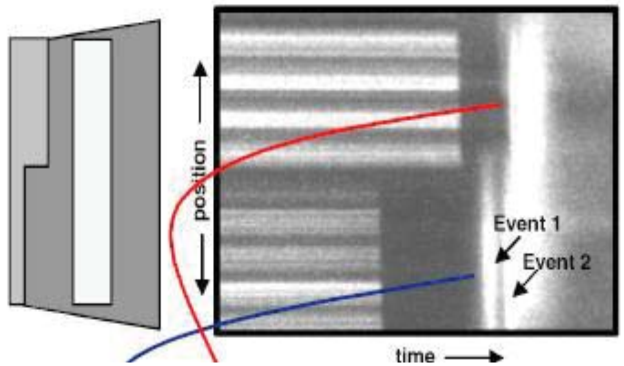


USP laser

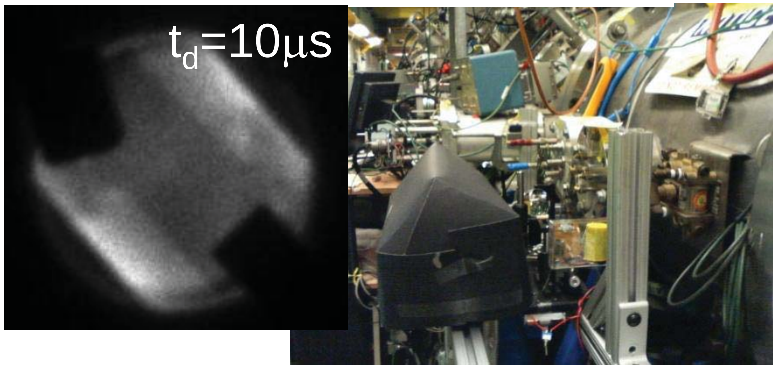


USP Ion beam

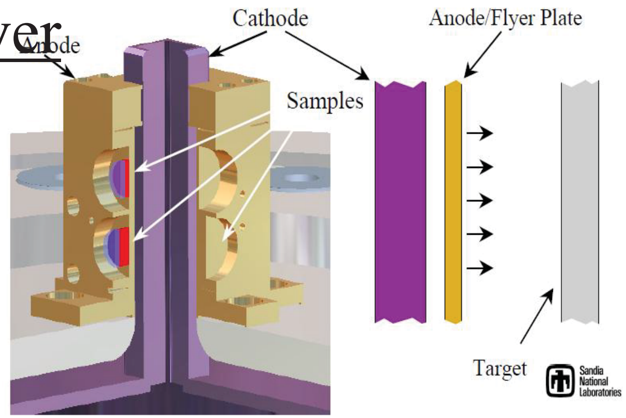
Laser shock (solid and form)



Accelerator Ion beam

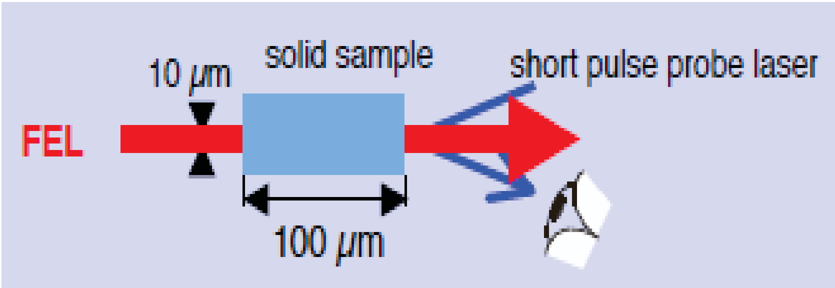


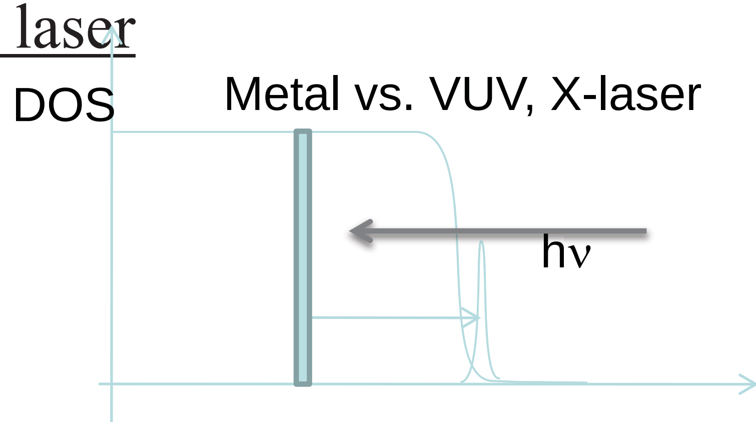
Flyer

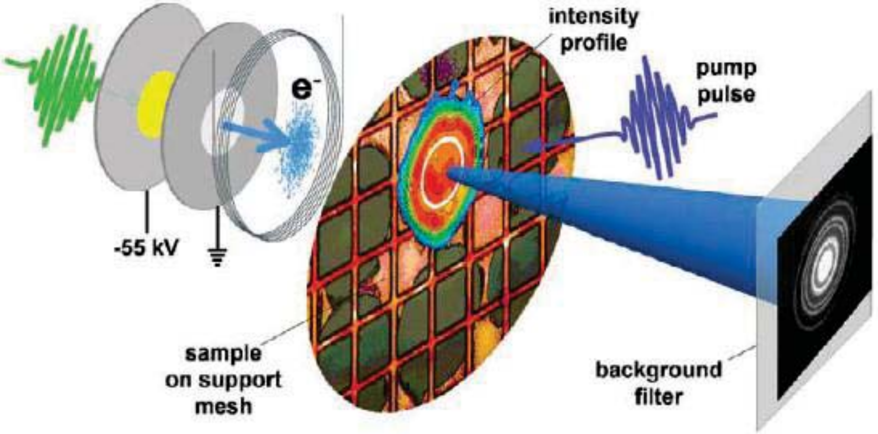


Recent new WDM experiments

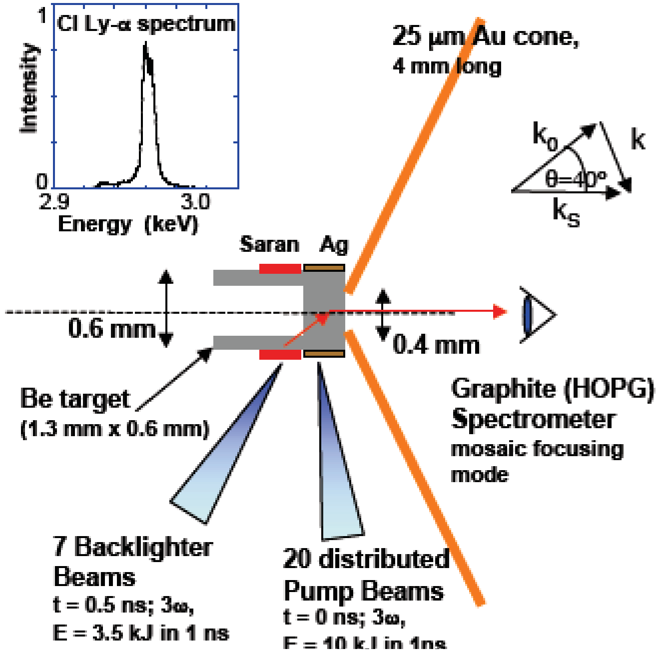
WDM created by VUV or X ray laser





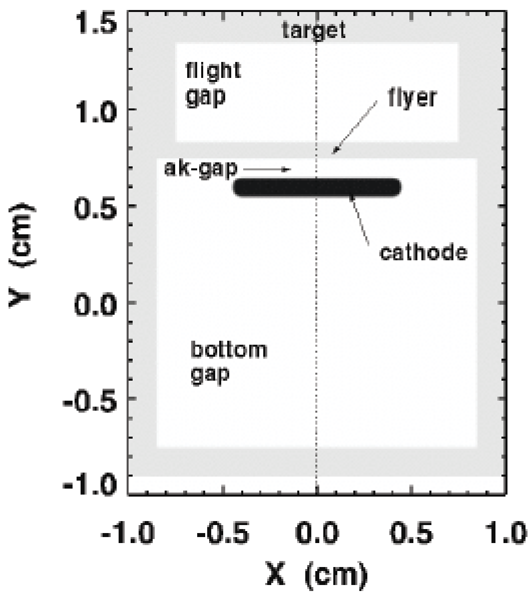


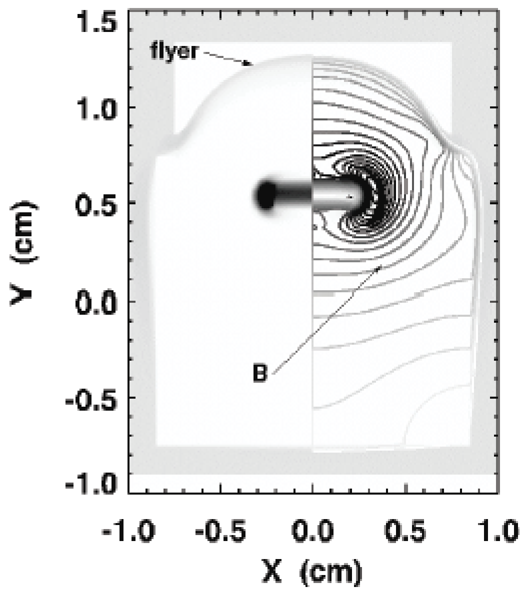
Pulsed e-beam diffraction

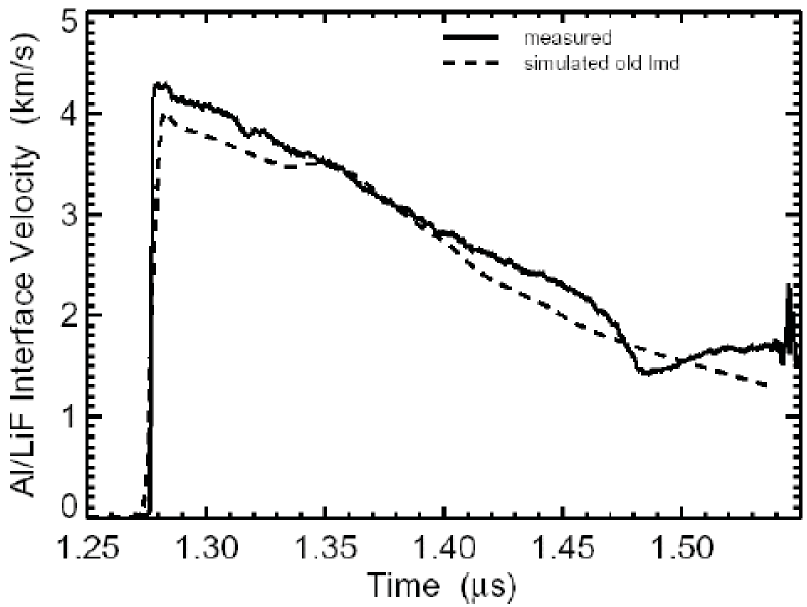


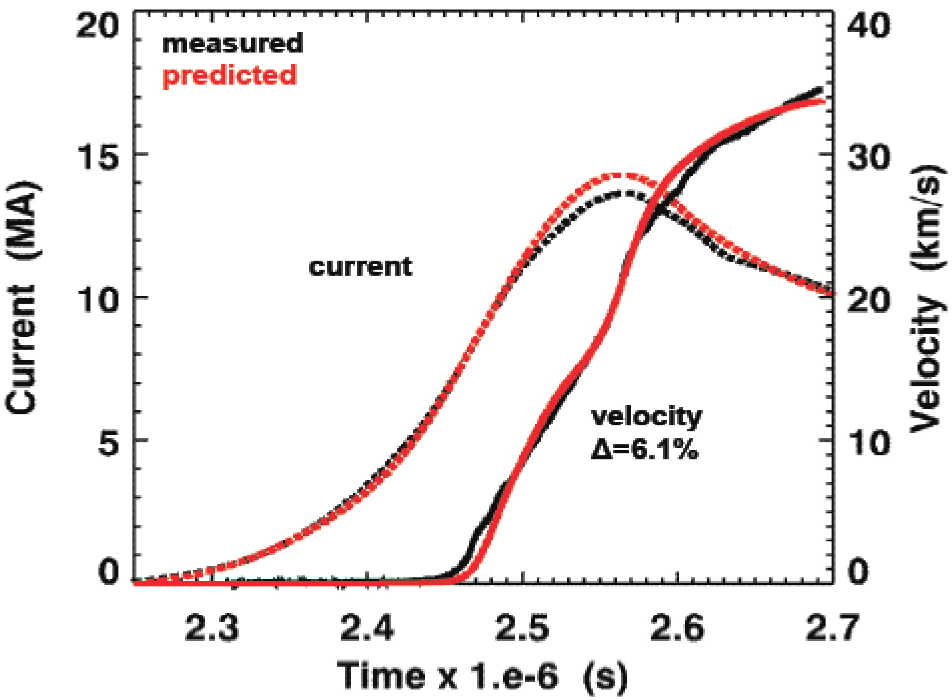
X-ray Thomson scattering

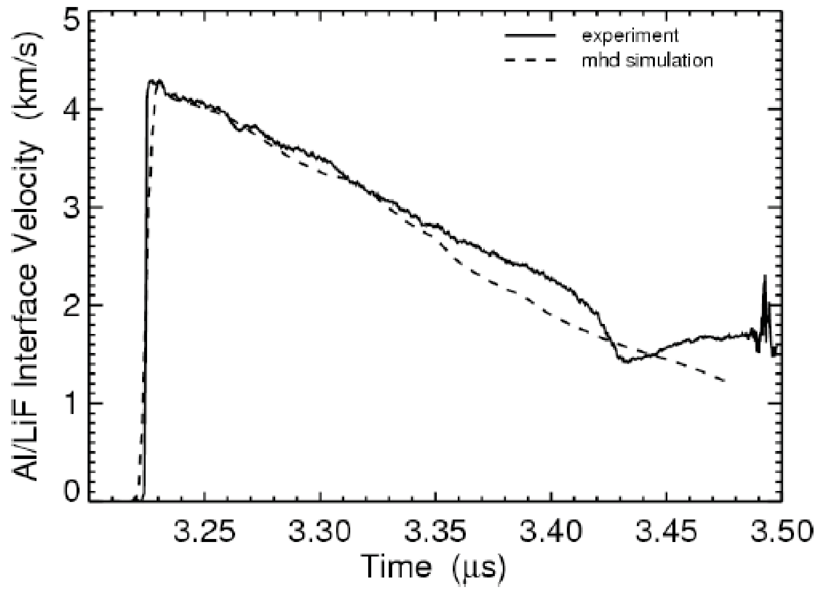
More accurate model we need
(SNL Flyer experiments)











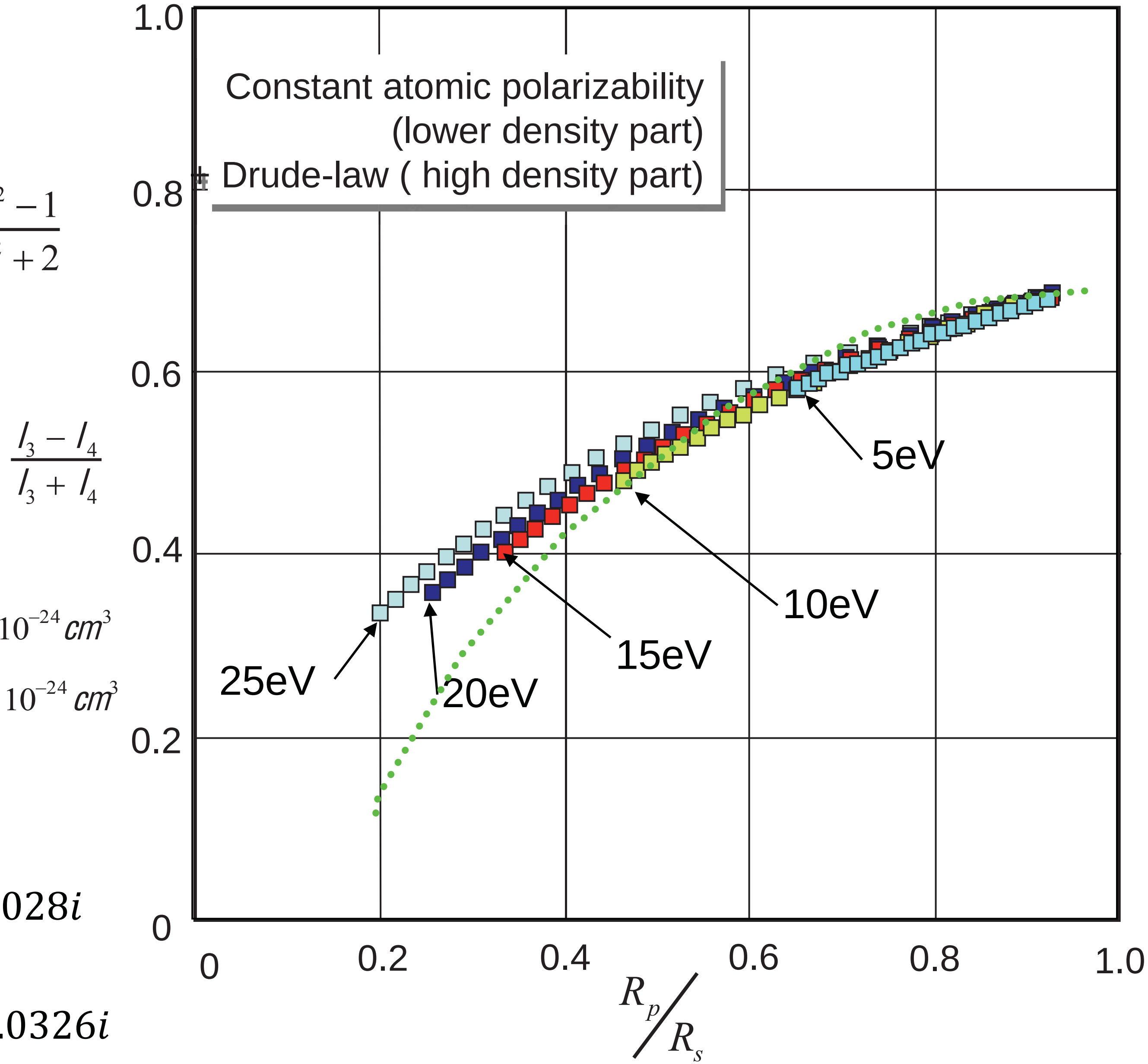
M. D 2008 WDM school

From USP laser measurement

Dense vapor of heated Au looks dielectric

$$P = Np = N\alpha E'$$

$$\alpha = \frac{3}{4\pi N}\frac{\varepsilon-1}{\varepsilon+2} = \frac{3}{4\pi N}\frac{n^2-1}{n^2+2}$$



$$\alpha(vapor) = (-1.75 + 0.2i) \times 10^{-24}\, cm^3$$

$$\alpha(metal) = (-29.4 + 0.14i) \times 10^{-24}\, cm^3$$

$$\varepsilon_{cold} = -6.38 + 0.028i$$

↓

$$\varepsilon_{heated} = 0.105 + 0.0326i$$

Fails

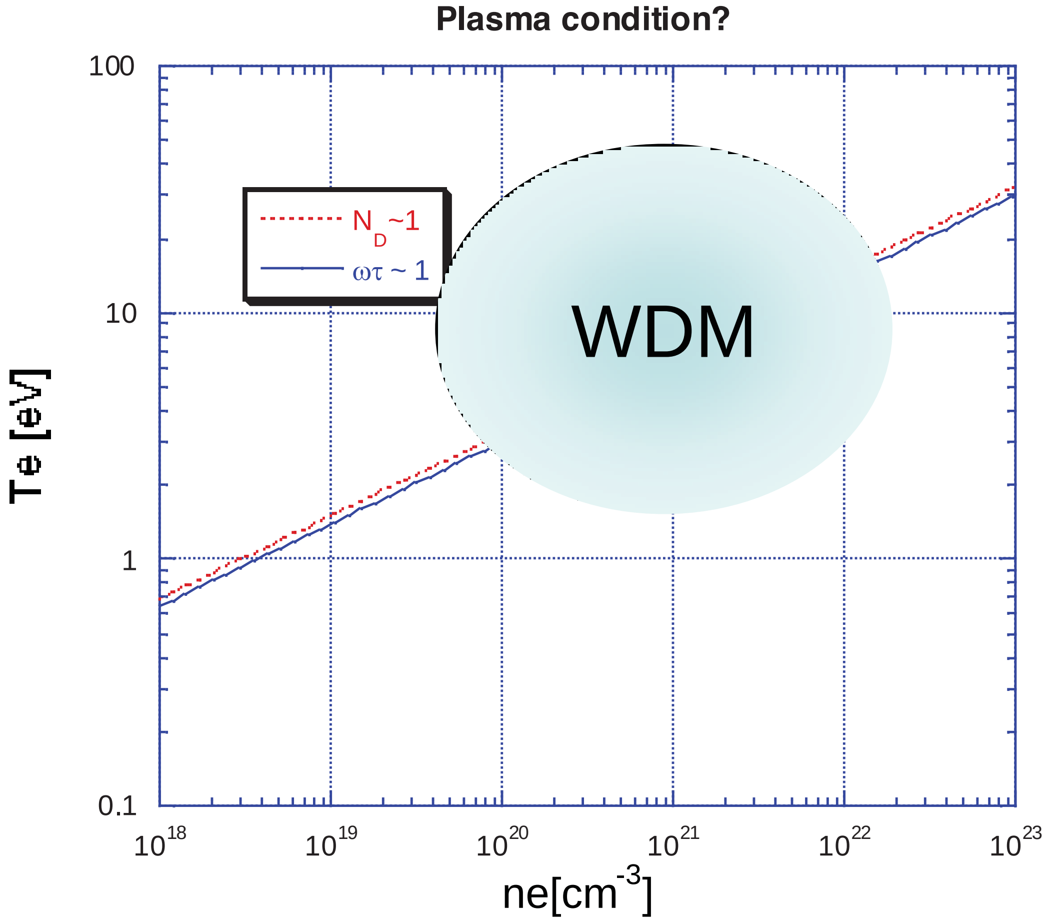
in the theoretical prediction from ordinary physics

Introduction of Plasma Physics
F. F. Chen

1.6 CRITIERIA FOR PLASMAS

….“The three conditions a plasma must satisfy are therefore: ”…

1. $\lambda_D << L$.
2. $N_D >>>1$.
3. $\omega\tau > 1$.



What should we consider in WDM condition?

Condensed matter side

- Energy band structure change
- Ordinary and disordinary
- electron localization
- Fermi surface change
- Atomic vibration and sound wave in (pseudo)lattice
- Debye model Temperature and density dependence
- Gruneien law, Cv
- Lindemann law, m.p. b.p.
- Latent heat of melting and boiling
- Metal-nonmetal transition
- Critical point

Plasma physics side

- Many body interaction
- Fermi degeneracy
- Equation of States
- Surface tension, viscosity
- two-phase fluids
- Electron affinity and negative-positive plasma
- Quantum effect in plasmas
- Collision model
- Nonequilibrium
- Screening
- Coulomb Logarithm
- Strongly coupled plasma

Chemical reaction side

- Reaction rate for dissociation and combination
- Equation of States
- Liquid-vapor interface
- Van der Waals' model
- Electron affinity
- Clusters
- Density functional theory

What should we consider in WDM condition?

Condensed matter side

- Energy band structure: change due to the excitation electrons
- Lattice: Ordinary => disordinary
- Electron: localization and delocalization
- Fermi surface: change and statistics also change (F-D => M-B, finally)
- Atomic vibration and sound wave: => plasmon? Ion wave?
- Debye model: => fail of the shielding model
- Gruneien law, Cv: => T dependence is change
- Lindemann law, m.p. b.p.: => pressure effect, density effect
- Latent heat of melting and boiling: =>change of number of freedom, heat bath is changed
- Metal-nonmetal transition: => Impurity? disordering?
- Critical point: we need accurate data.
- Density functional theory: excitation states should be included.

What should we consider in WDM condition?

Chemical reaction side

- dissociation and combination: n & T dependence of the rate
  (molecular dissociation at $T \sim 10^4 K$)
- Equation of States: phase transition? Two fluid(gas & liquid)
- Liquid-vapor interface: more energetic particle
- Van der Waals' model: We may need to include excitation energy.
- Electron affinity: T and n dependence?
- Clusters: highly charged?

What should we consider in WDM condition?

Plasma physics side

- Many body interaction
- Strongly coupled
- Fermi degeneracy
- Equation of States
- Surface tension, viscosity
- two-phase fluids
- Electron affinity with ionization and excitation
- Quantum effect
- Collision model and screening effect
- Non-equilibrium
- Coulomb Logarithm
- ………………………

Why do the main theories of matter fail at WDM conditions?

Condensed matter

T increase

Solid => liquid =>ionizing
Atoms in solid are no longer neutral
and no longer have ground state conditions.

Dense chemistry

N increase

Pressure effect on molecules
Reaction rate at high density and high temperature

$N + N \Rightarrow N_2$ $k=10^{15} cm^6 mol^{-2} s^{-1}$ If $[N] \sim 10^{22} cm^{-3}$, $t_{reaction} \sim 3.6ps$!?$<\tau_{ii}$

Low temperature plasma

N increase, T decrease

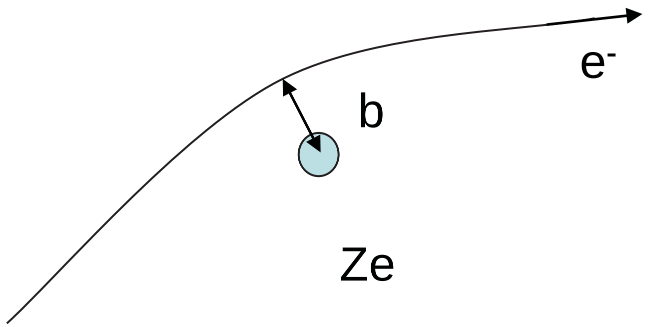
$\lambda_D > L$, $N_D < 1$, $\omega\tau < 1$
Neutral atoms and molecules dominate ions

What, how, why, where

For example,

- Extrapolation for present physical theory and checking.
- New possibility for applications
- Creation general model and/or parameter data base
- Construction of physical model with similarity to extreme condition
- Reconsider the present modeling

Coulomb Logarithm



$$\Delta v = \frac{Ze^2}{mb^2}\left(\frac{2b}{v}\right)$$

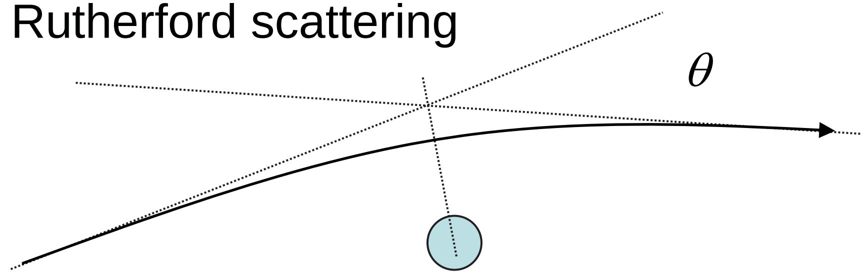
$$\frac{d}{dt}\left\langle (\Delta v)^2 \right\rangle = \int 2\pi b\, db\, n_i v (\Delta v)^2$$

$$\left\langle (\Delta v)^2 \right\rangle = \frac{8\pi n_i Z^2 e^4 \ln \Lambda}{m^2 v}$$

$$\Lambda = \frac{b_{\max}}{b_{\min}} \qquad b_{\min} = \frac{Ze^2}{mv^2}\ or\ \frac{\hbar}{mv}$$

$$b_{\max} = \lambda_{Debye} = \left(\frac{\varepsilon_0 k_B T_e}{n_0 e^2}\right)^{1/2}$$

Rutherford scattering



$$\sigma_{tr} = \int \frac{d\sigma}{d\Omega}(1-\cos\theta)\, d\Omega$$

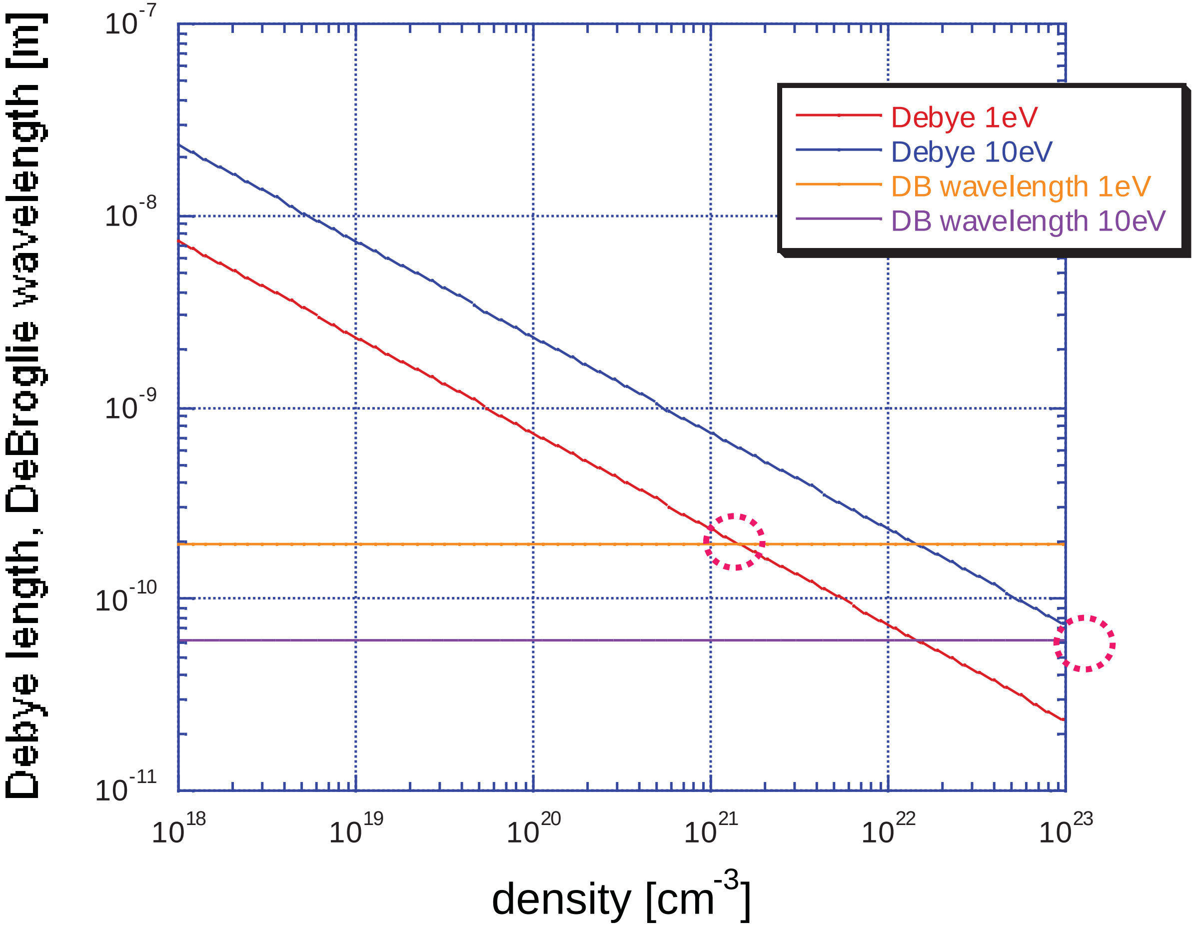
$$= \frac{4\pi Z^2 e^4}{m^2 v^4}\int \frac{d(\sin\theta/2)}{\sin\theta/2} = \frac{4\pi Z^2 e^4}{m^2 v^4}\log\left(\frac{2}{\theta_{\min}}\right)$$

When smaller T and larger n,
then b_{max} is sometimes comparable with b_{min}.

Fail in Debye screening model
=>We can't define b_{max}, θ_{min}.

When does it fail?

$b_{min} > b_{max}$



M.S.Murillo, POP, Vol.11-5, p.2964 (2004)

From theory of one component plasma (shielding)

$$-\varepsilon_0 \nabla^2 \phi(r) = Qn(r) - Qn$$ Poisson equation

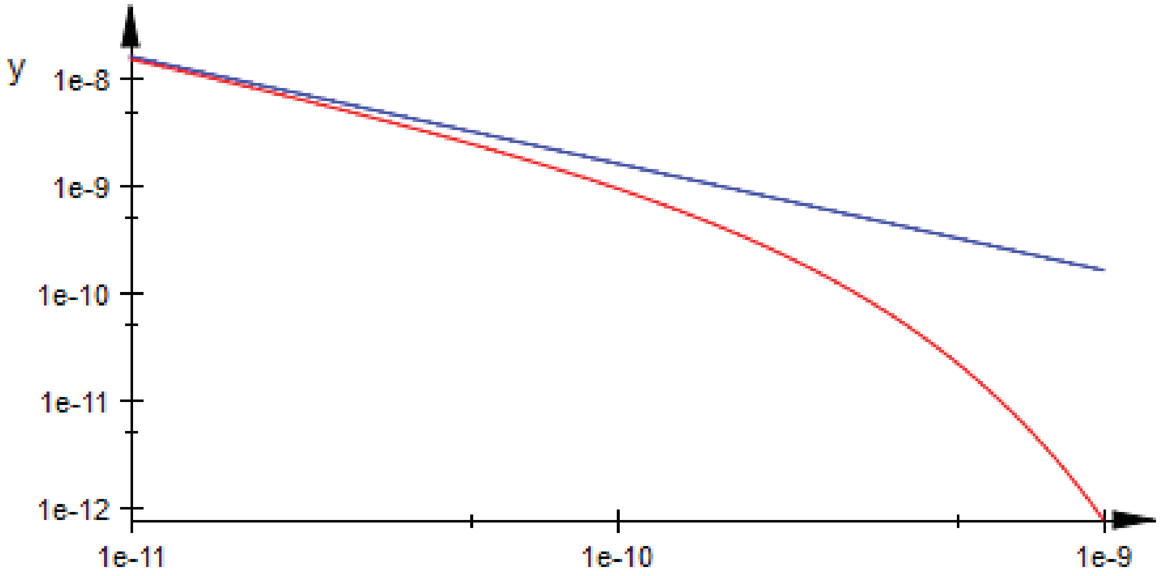
$$n(r) = n \cdot exp\left(-\frac{Q\phi(r)}{T}\right) \approx n\left(1 - \frac{Q\phi(r)}{T}\right)$$ Boltzmann law

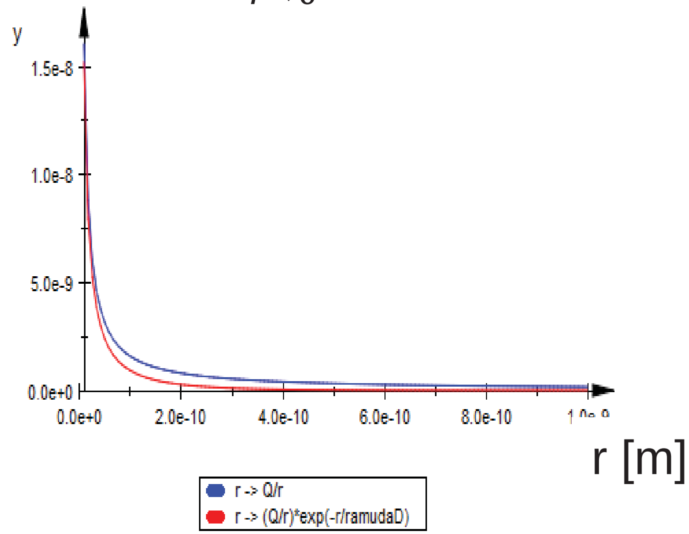
$\frac{Q\phi(r)}{T} << 1$

$$\phi(r) = \frac{1}{4\pi\varepsilon_0}\frac{Q}{r}$$

$$\lim_{r\to 0} r\phi(r) = \frac{Q}{4\pi\varepsilon_0}$$

$$\phi(r) = \frac{Q}{4\pi\varepsilon_0 r} exp\left(-\frac{r}{\lambda_D}\right) \quad \left(\lambda_D = \sqrt{\varepsilon_0 T/Q^2 n}\ \right)$$





Te = 1eV, ne=nc(λ=1μm, 1.6 x 10^{21}cm^{-3})

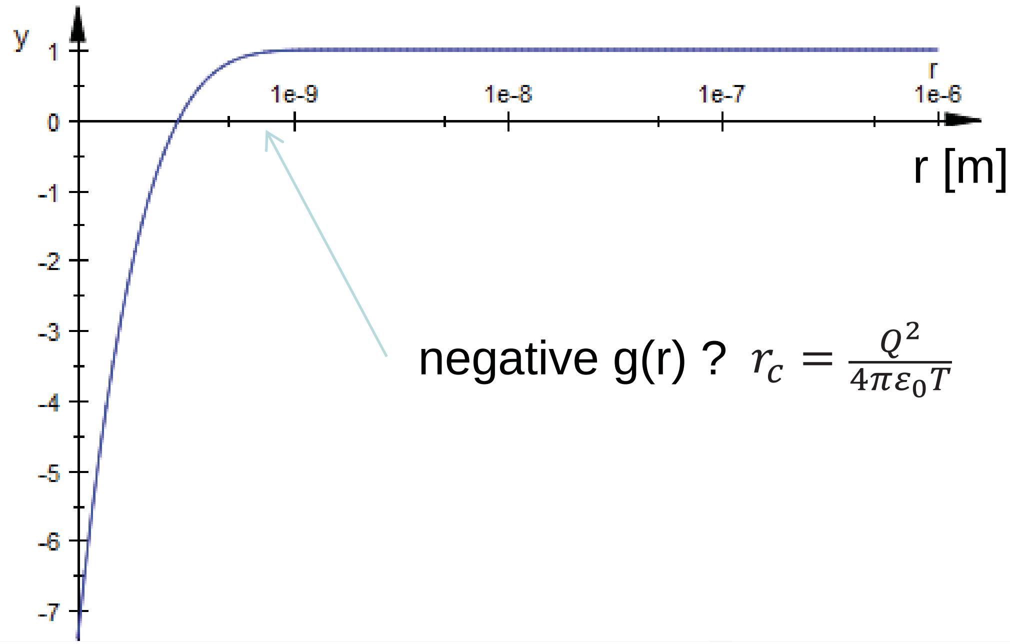
r -> Q/r
r -> (Q/r)*exp(-r/ramudaD)

Debye-Huckel model

Debye shielding

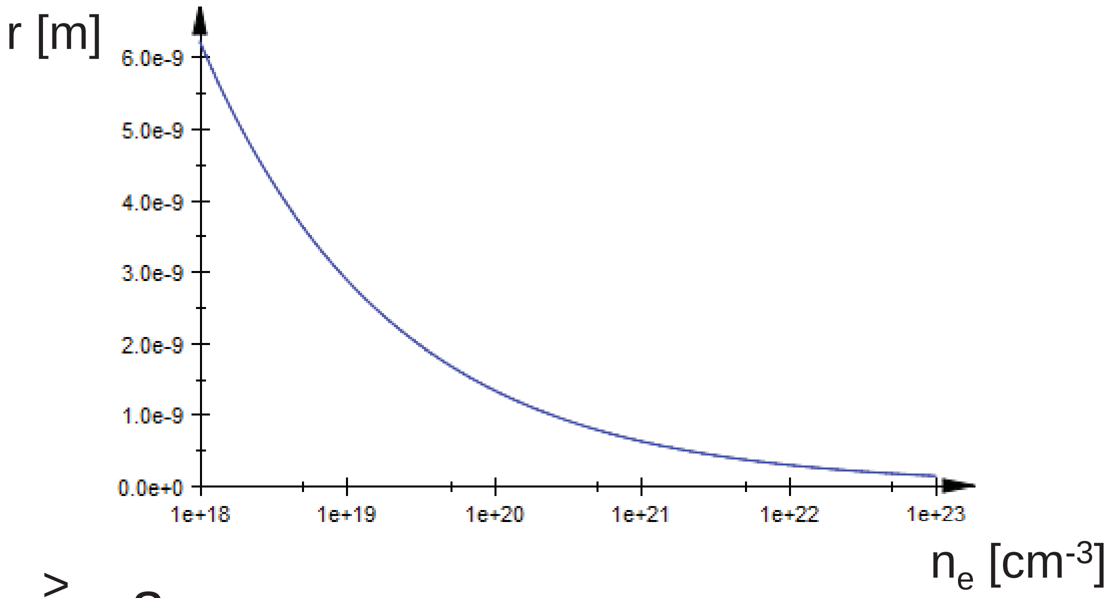
Distribution function

$$g(r) = \frac{n(r)}{n} = \left[1 - \frac{Q^2}{4\pi\varepsilon_0 T r} exp\left(-\frac{r}{\lambda_D}\right)\right]$$



Radius of ion sphere

$$a = (3/4\pi n)^{1/3}$$



$$r_c \lesseqgtr a$$

Question: why?

Treatment of transport of electrons is different .

$$J = ne\mu_e E$$

$$\tau : relaxation\_time$$

$$m^*\left(\frac{dv}{dt} + \frac{v}{\tau}\right) = -eE$$

$$\mu_e = \frac{e\tau}{m^*}$$ (d/dt=0)

from textbook of Semiconductor phys.

$$m\ddot{\mathbf{r}} + m\beta\dot{\mathbf{r}} = e\mathbf{E}$$

$$\mathbf{r} = -\frac{e}{m(\omega^2 + i\beta\omega)}\mathbf{E}$$

$$\mathbf{j} = Ne\dot{\mathbf{r}} = \frac{Ne^2}{m(\beta - i\omega)}\mathbf{E}$$

$$\sigma = \frac{Ne^2}{m(\beta - i\omega)}$$

$$\mu\hat{\varepsilon} \equiv \hat{n}^2 = \mu\left(\varepsilon + i\frac{4\pi\sigma}{\omega}\right)$$

$$\hat{\varepsilon} = 1 - \frac{4\pi Ne^2}{m}\frac{1}{\omega(\omega - i\beta)}$$

from textbook of electromagnetism

$$m\ddot{\mathbf{r}} + q\mathbf{r} = e\mathbf{E}$$ $+ g\dot{\mathbf{r}}$

$$\mathbf{r} = -\frac{e}{m(\omega^2 - \omega_0^2)}\mathbf{E}, \_ \omega_0^2 = \frac{q}{m}$$

$$\mathbf{P} = N\mathbf{p} = Ne\mathbf{r} = \frac{Ne^2}{m(\omega_0^2 - \omega^2)}\mathbf{E}$$ $-i\omega g$

$$= N\alpha\mathbf{E}$$

$$\eta = \frac{N\alpha}{1 - \frac{4\pi}{3}N\alpha}$$

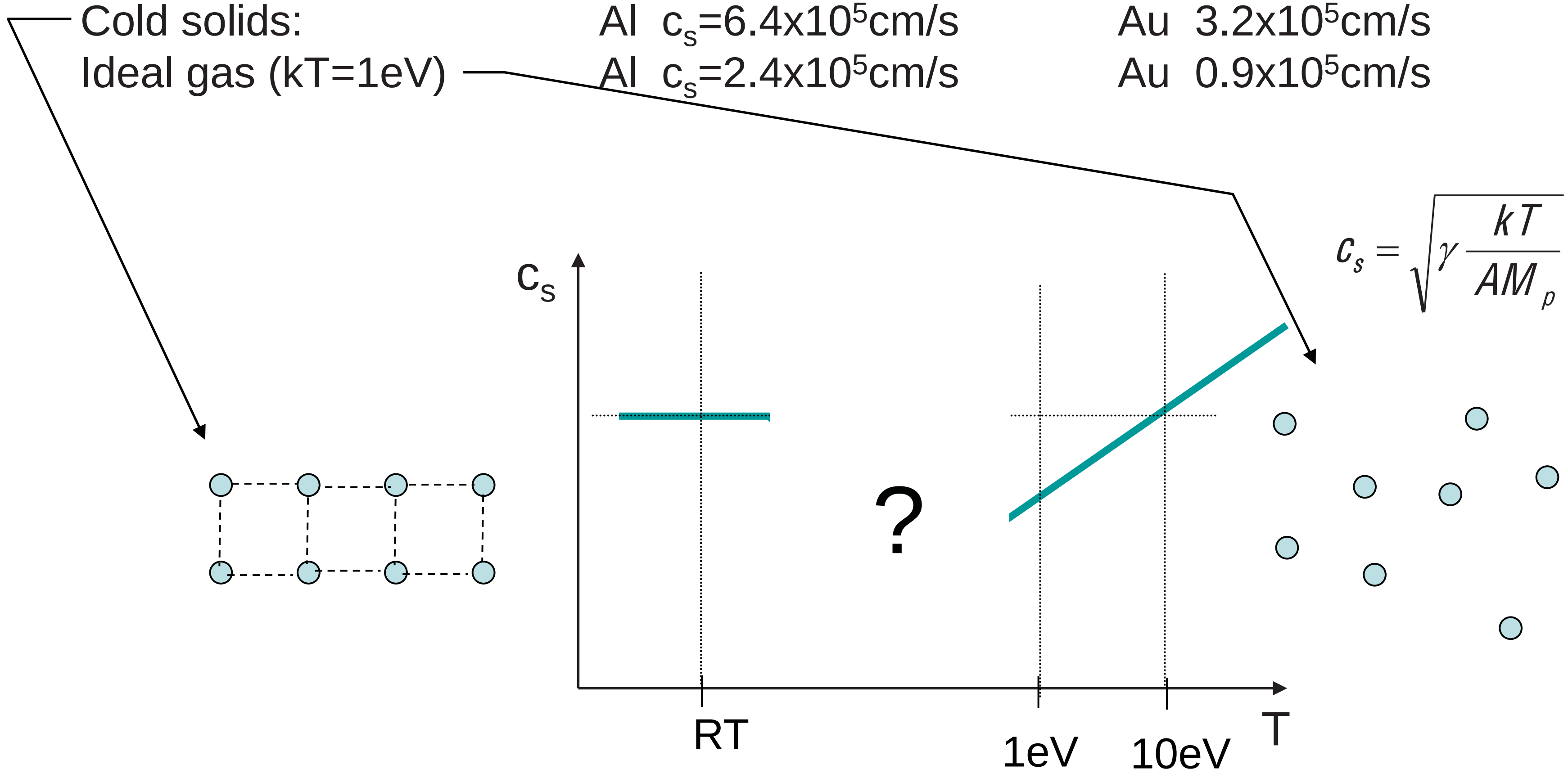
$$\varepsilon = \frac{1 + \frac{8\pi}{3}N\alpha}{1 - \frac{4\pi}{3}N\alpha}$$

from textbook of optics

Knowledge from known physics

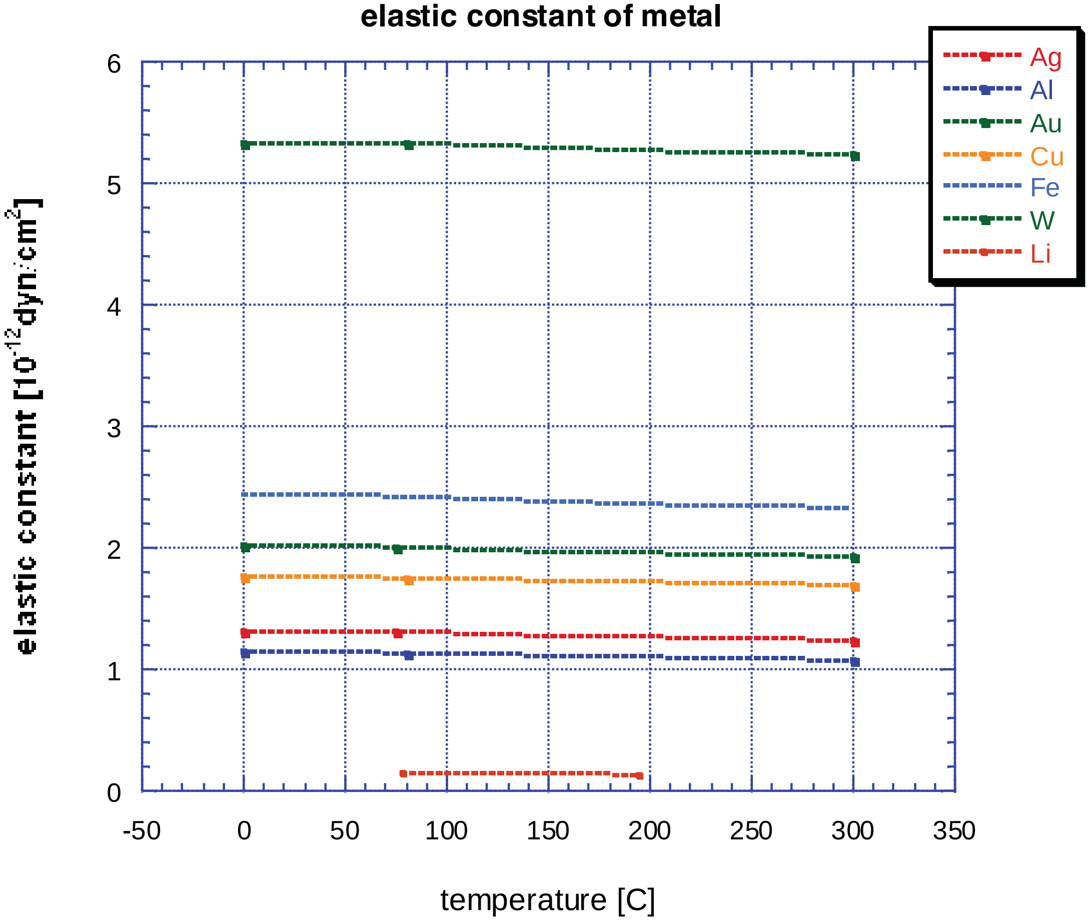
Sound velocity

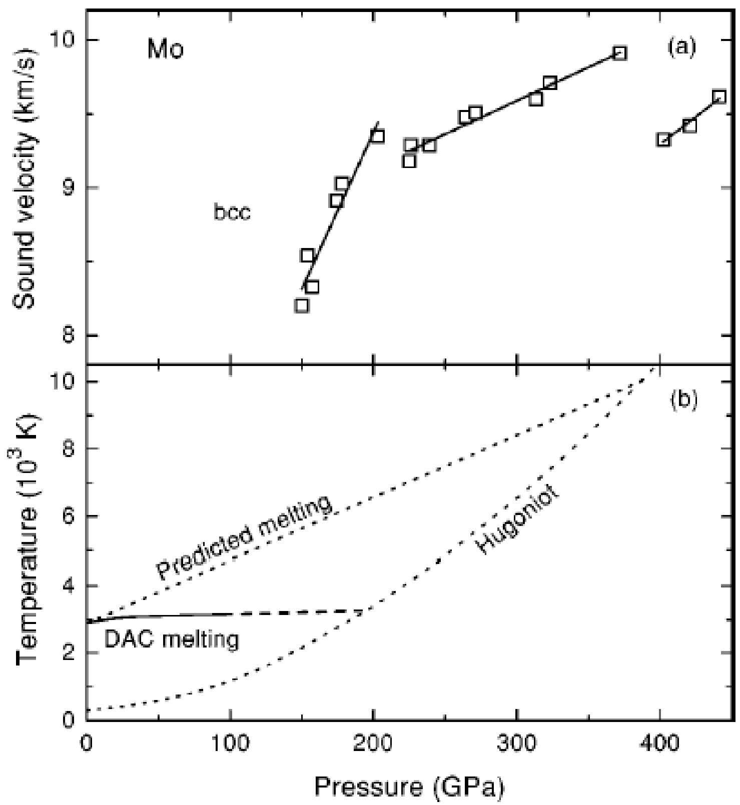
Cold solids:	Al c_s=6.4x10^{5}cm/s	Au 3.2x10^{5}cm/s
Ideal gas (kT=1eV)	Al c_s=2.4x10^{5}cm/s	Au 0.9x10^{5}cm/s



$$c_s = \sqrt{\gamma \frac{kT}{AM_p}}$$

Temperature dependence of most metals are negligible small.





Daniel Errandonea,
PHYSICAL REVIEW B, VOLUME 63, 132104

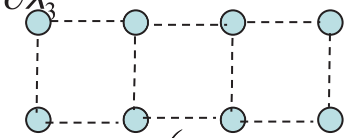
Sound velocity in metal = elastic modulus/density
(Normally, elastic modulus has small dependence with temperature.)

Sound wave in metals (Lattice)

Motion equation

$$F_1 = \left(\frac{\partial T_{11}}{\partial x_1} + \frac{\partial T_{12}}{\partial x_2} + \frac{\partial T_{13}}{\partial x_3} \right) dx_1 dx_2 dx_3$$

$$\rho \frac{\partial^2 u_1}{\partial t^2} = \frac{\partial T_{11}}{\partial x_1} + \frac{\partial T_{12}}{\partial x_2} + \frac{\partial T_{13}}{\partial x_3}$$



$$\rho \frac{\partial^2 u_i}{\partial t^2} = \frac{\partial T_{ij}}{\partial x_j}$$

$$S_{ij} = \frac{1}{2}\left(\frac{\partial u_k}{\partial x_l} + \frac{\partial u_l}{\partial x_k} \right)$$

Hook's law

$$\rho \frac{\partial^2 u_1}{\partial t^2} = c_{11} \frac{\partial^2 u_1}{\partial x_1^2}$$

$$T_{ij} = c_{ijkl} S_{kl}$$

$$u_1 = u_0 \exp(ikx_1 - i\omega t)$$

$$\omega = c_s k$$

$$c_s = \sqrt{c_{11} / \rho}$$

Sound wave in metals (free electrons)

$$V_s = \left(\frac{mZ}{3M} \right)^{1/2} V_{Fermi} = \left(\frac{mZ}{3M} \right)^{1/2} \frac{\hbar}{m} \left(3\pi^2 n_e \right)^{1/3}$$

Velocity on Fermi surface

Sound wave in plasmas

Fluid equation of ion fluid

$$Mn\left[\frac{\partial v_{ion}}{\partial t} + (v_{ion} \cdot \nabla) v_{ion} \right] = enE - \nabla p = -en\nabla\phi - \gamma_{ion} k_B T_{ion} \nabla n$$

Plane wave approximation

$$-i\omega M n_0 v_{i1} = -en_0 ik\phi_1 - \gamma_1 k_B T_i k n_1$$

electron: m=0 quick equilibrium with potential ϕ

$$n_e = n = n_0 \exp\left(\frac{e\phi_1}{k_B T_e} \right) \approx n_0 \left(1 + \frac{e\phi_1}{k_B T_e} + \cdots \right)$$

$$\therefore \quad n_1 = n_0 \frac{e\phi_1}{k_B T_e}$$

Continuum equation of ions

$$i\omega n_1 = n_0 ik v_{i1}$$

$$i\omega M n_0 v_{i1} = \left(en_0 ik \frac{k_B T_e}{en_0} - \gamma_1 k_B T_i ik \right) \frac{n_0 ik v_{i1}}{i\omega}$$

$$\omega^2 = k^2 \left(\frac{k_B T_e}{M} + \frac{\gamma_1 k_B T_i}{M} \right)$$

$$c_s = \frac{\omega}{k} = \sqrt{\frac{k_B T_e + \gamma_1 k_B T}{M}}$$

Ion density fluctuation

What parameters decided material condition?

Can you image what’s materials?

material	N_2	Na	Al
Solid density	1.03 g/cc	0.97 g/cc	2.68 g/cc
Ionization potential(atom)	14.5eV	5.14eV	5.99eV
IP for molecule	15.6eV	5.14eV	??
Cohesive energy of solid	4.9eV/atom	1.11eV/atom	3.39eV/atom
Interatomic distance	1.76 Å	2.08 Å	1.6 Å
Melting temperature	63.16K	371K	933K
Boiling temperature	77.36K	1156K	2792K

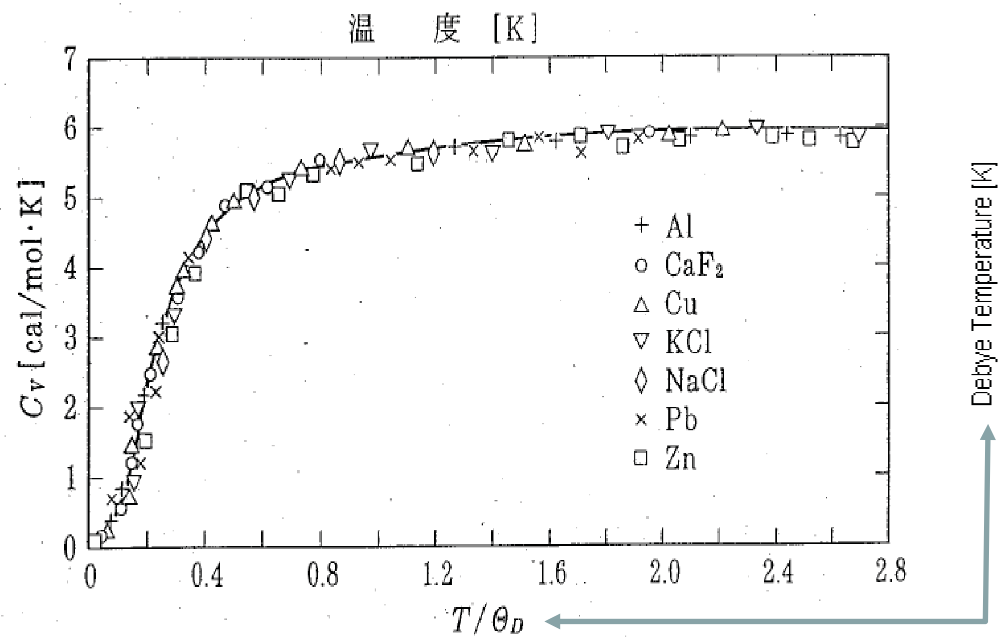
Question: why?

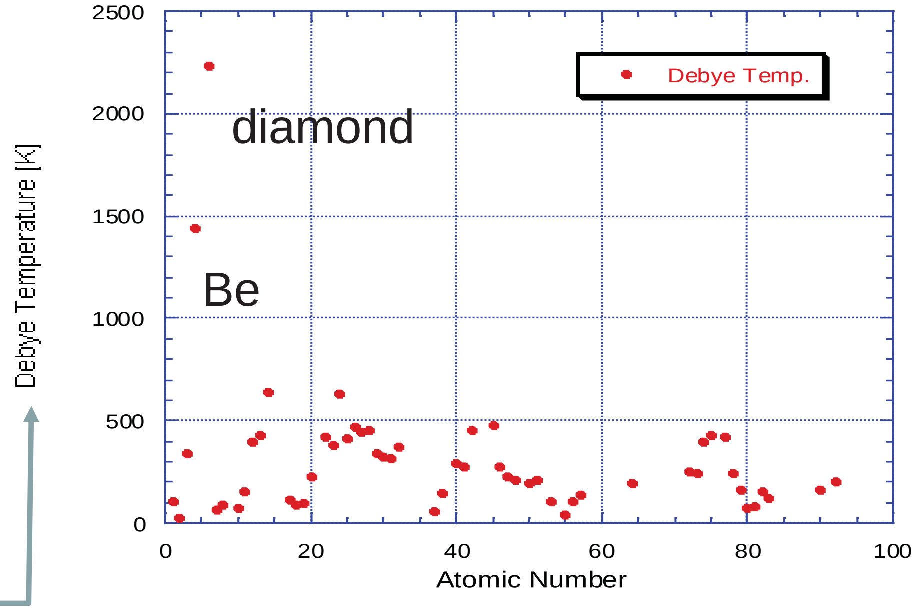
It is not enough to explain the material condition with solid density, ionization energy, cohesive energy, interatomic distance.

What is general feature of condensed matter?

The Debye model for C_v agrees well with experimental data

$$c_v = 3Rf_D(\Theta_D / T) = 3R(T/\Theta_D)^3 \int_0^{\Theta_D/T} \frac{e^x x^4}{(e^x - 1)^2} dx$$





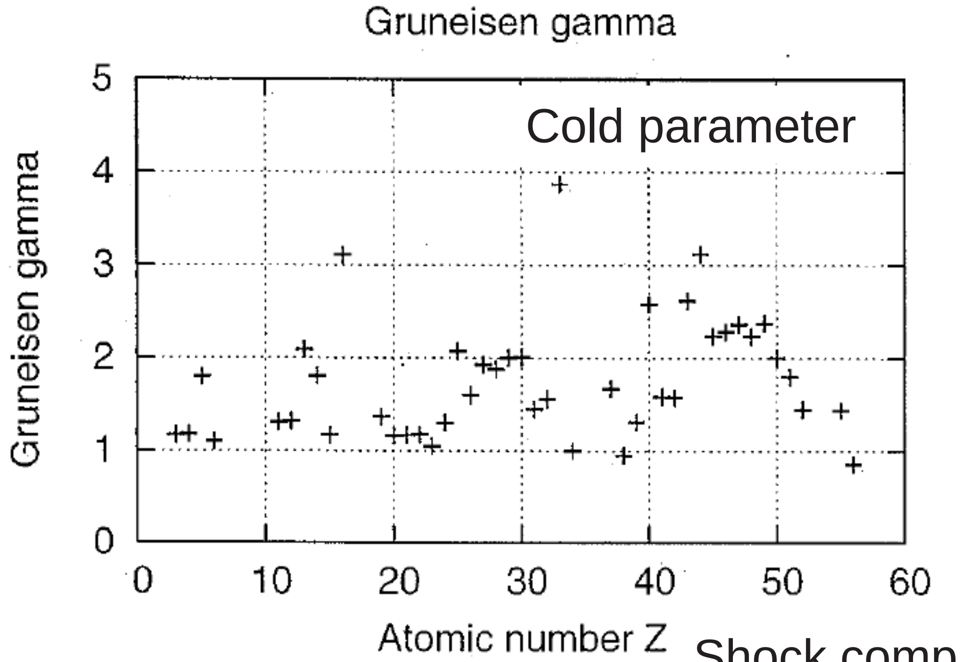
The effect of thermal expansion or compression
=>Gruneisen parameter

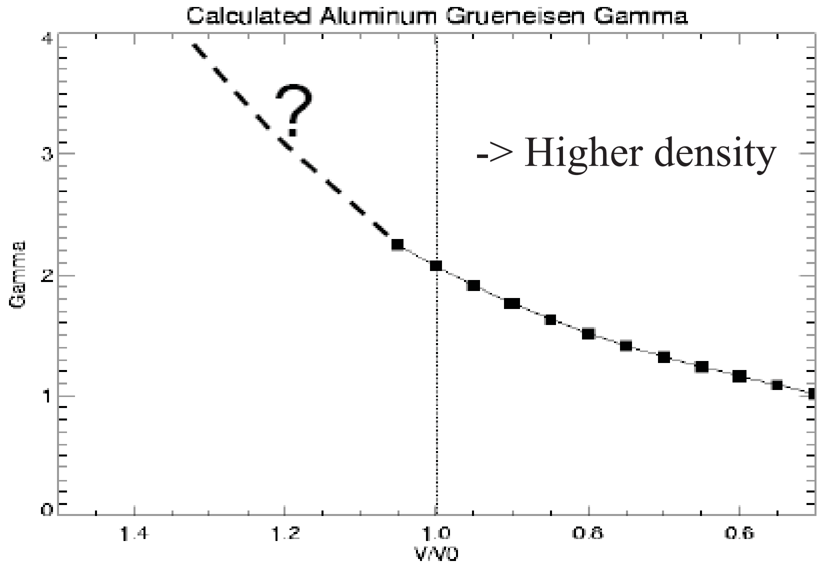
α : thermal_expansion_coefficient

χ : Compressibility

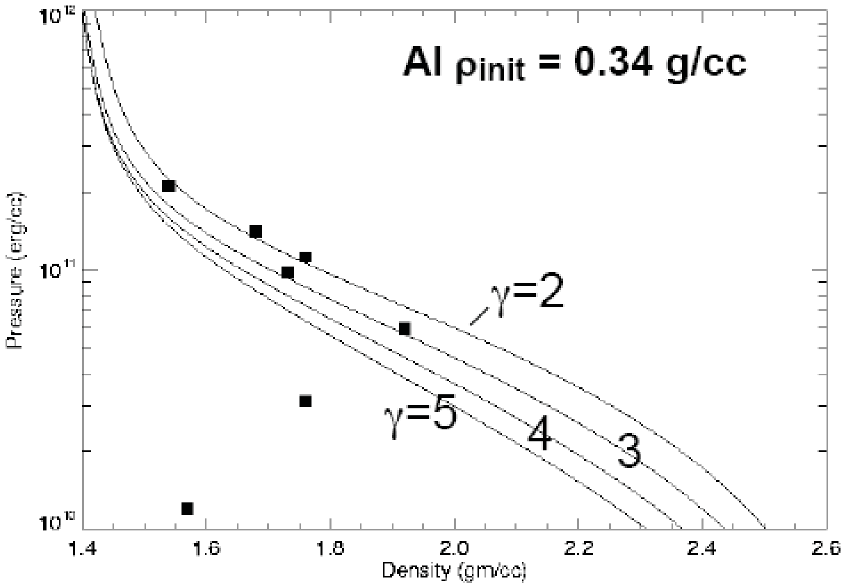
$$\gamma_0 = \frac{\alpha V_0}{\chi C_v} = \frac{d(\ln \Theta_D)}{d(\ln \rho)}$$

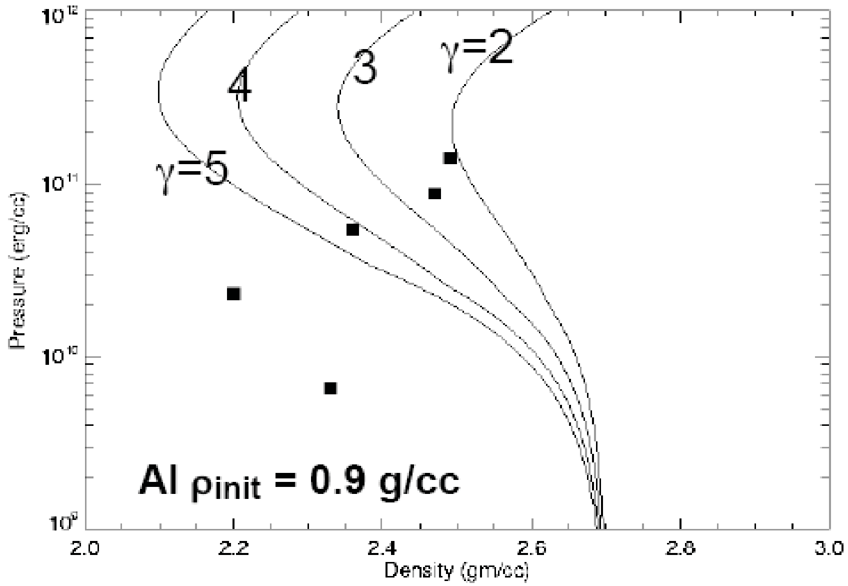
Dependence of Gruneisen parameter and effect on the EOS



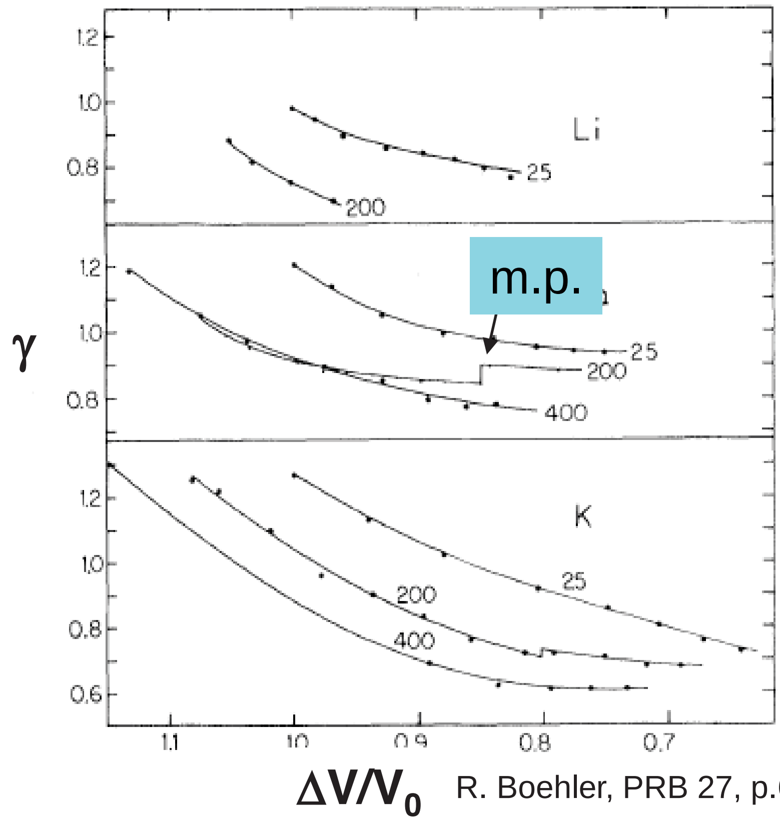


Shock compression results





More details of Gruneisen parameter in condensed matter



R. Boehler, PRB 27, p.6754

general feature

$$\gamma = \gamma_0 \left(V / V_0 \right)^q$$

Cu $q=1.08\sim1.33$

Fe $q=0.6\sim0.783$

But, finally details of electron orbits are needed.

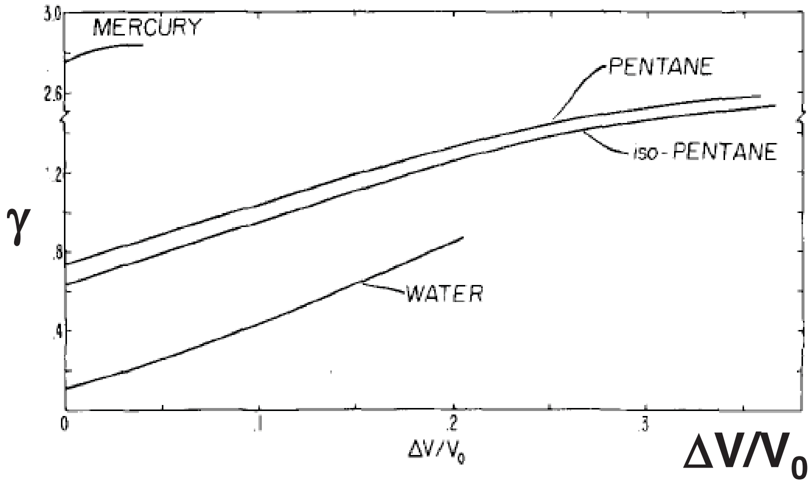
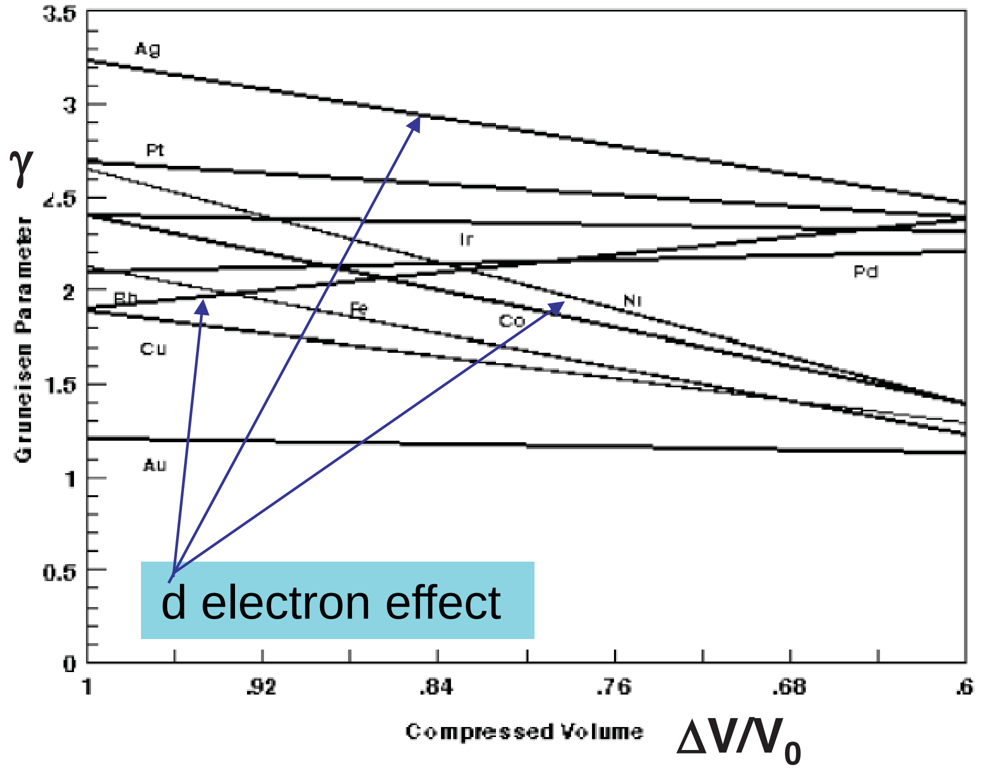


FIG. 5. Volume dependence of the Grüneisen parameter of water, isopentane, pentane, and mercury at room temperature.

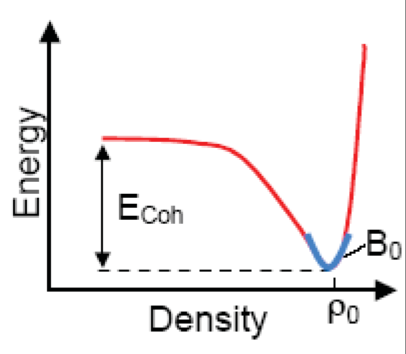
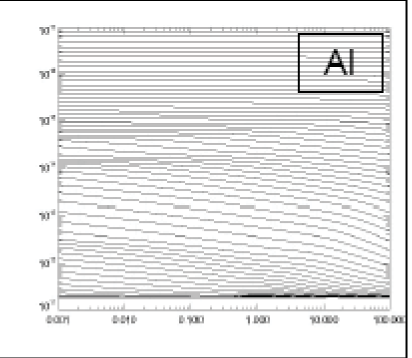
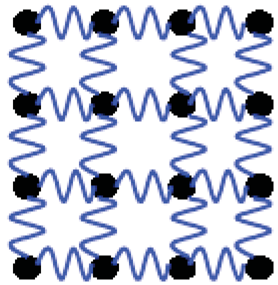
R. Boehler, JAP 48, p.4183



C. V. Pandya, Bull. Mater. Sci, 25, p.63 (2002)

How to consider EOS model in WDM?

$$F[\rho,T] = E_{Cold}[\rho] + F_{Elec}[\rho,T] + F_{Ion}[\rho,T]$$

Cold Curve	Electron-Thermal	Ion-Thermal
		
Cold curve may include additional parameters to modify $\rho < \rho_0$ and $\rho > \rho_0$ behavior	Ionization features seen in Purgatorio-based electron-thermal energy and entropy.	Ion thermal contribution from Debye model and density-dependent Grueneisen $\gamma[\rho]$

Sterne, WDM workshop in Pleasanton (2007)

i.e. for transition metals

$$E = E_i + E_s + E_{sd} + E_d + E_{ol} + \dots$$

Electrostatic interaction of ions core

energy of s-electrons interact with core and s-s

energy of d-electrons interact with core and d-d

hybrid term between s-d

energy due to the overlap of d states at different atomic sites

Rather simple but physically motivated model

$E = E_i + E_s^{(0)} + E_s^{(1)} + E_s^{(2)} + \Phi_{sr}$

$$E_s^{(0)} = 0.5\left(\frac{2.21}{r_s^2} - \frac{0.916}{r_s} + Z(-0.115 + 0.31 \ln r_s)\right) \qquad (2.7)$$

$$E_s^{(1)} = \lim_{q \to 0}\left(W(q) + \frac{4\pi Z}{Vq^2}\right), \qquad (2.8)$$

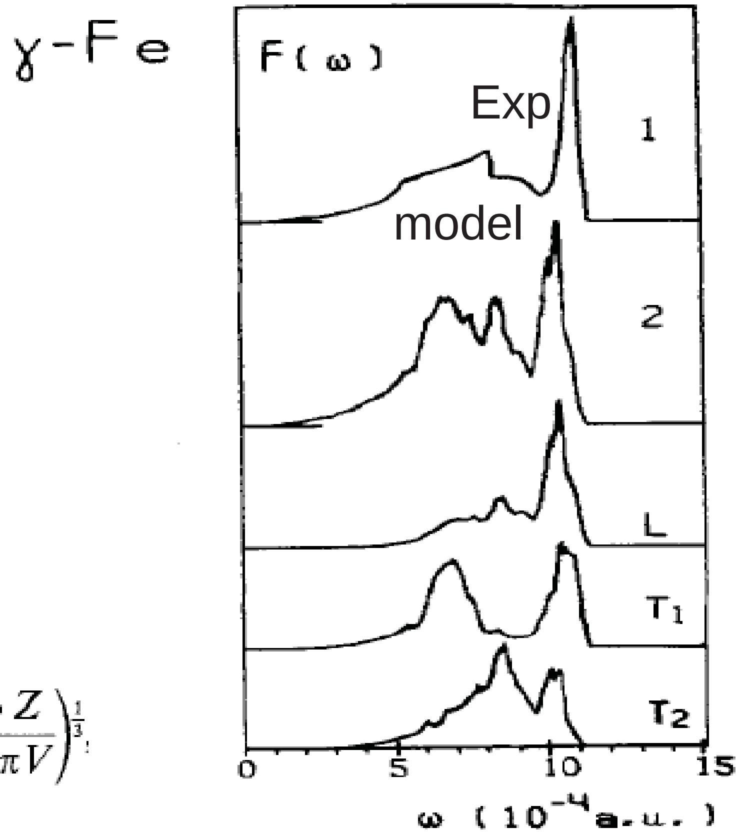
$$E_s^{(2)} = \sum_{\mathbf{G}} |S(\mathbf{G})|^2 F(G), \qquad (2.9)$$

$$F(q) = \frac{V}{2}\frac{\Pi(q)}{\varepsilon(q)} |W(q)|^2, \qquad (2.10)$$

$$W(q) = -\frac{4\pi Z}{Vq^2}\left[(1+U)\cos qR_0 - U\frac{\sin qR_0}{qR_0}\right] \cdot \exp\left(-0.03\left(\frac{q}{2k_f}\right)^4\right). \qquad (2.11)$$

$$\Phi_{sr} = 0.5\sum_{\mathbf{R}} V_{sr}(R) = 0.5\, A \sum_{\mathbf{R}} \exp(-\alpha R).$$

$$r_s = \left(\frac{3Z}{4\pi V}\right)^{\frac{1}{3}}$$



V.N. Antonov, Z Phys, B 79, 223

Lattice constant

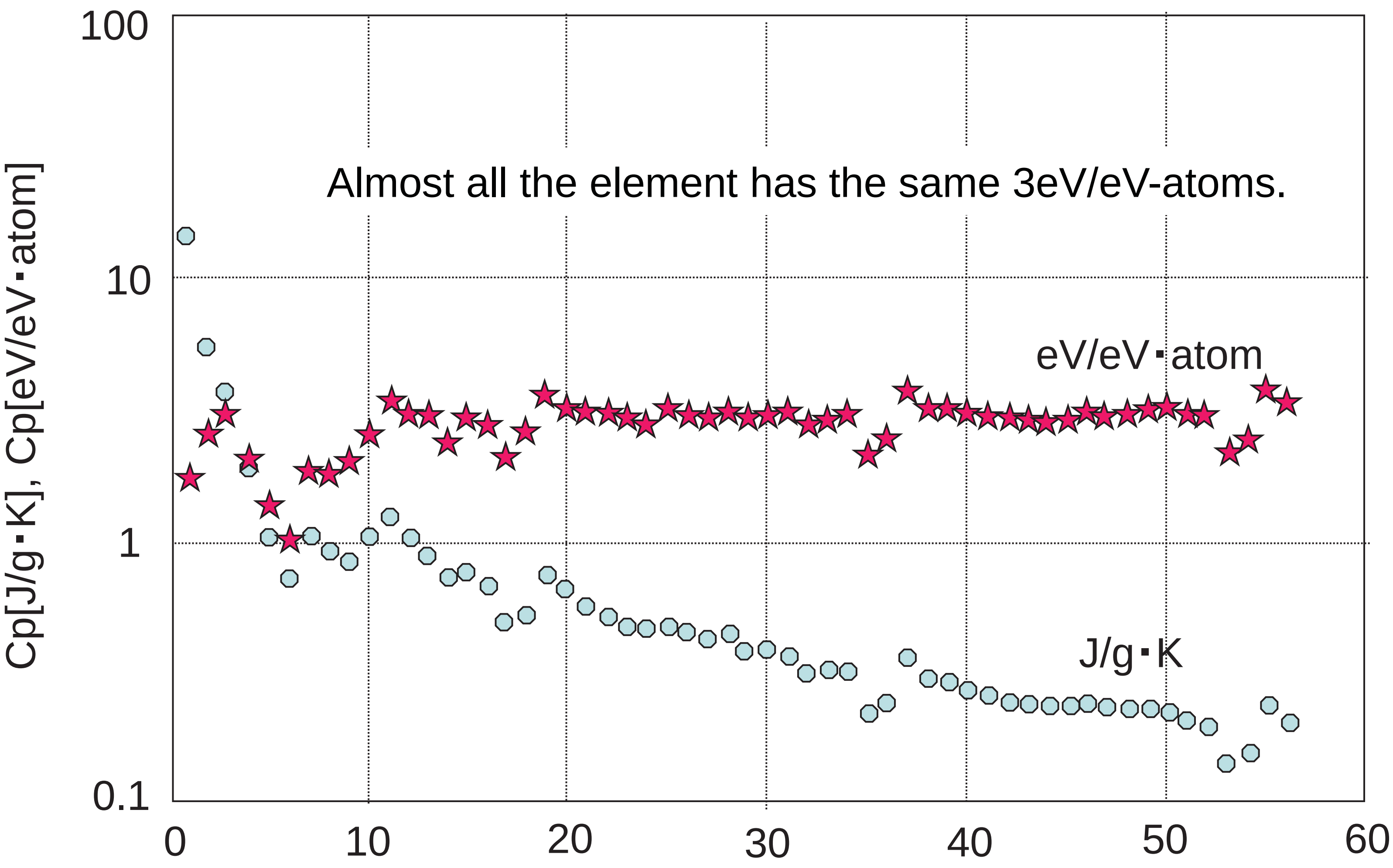
Correction factor for pseudopotential

Atom	a_0	U	R_0	A	α	$A' \cdot 10^3$	z
Cu	6.7973	−1.5074	2.7542	125.89	2.151	1.03	1
Ni	6.6592	−1.9379	2.5656	282.16	2.150	11.32	1
Co[a]	6.7048	−1.7812	2.5461	208.79	2.122	8.92	1
Fe[b]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Ag	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
Pd	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	1.6
Rh	7.1868	153634.0	0.0014	121284.0	3.482	2.51	1.9
Au	7.7076	−822.72	0.0500	258.80	1.800	14.21	1
Pt	7.4004	10947.2	0.0042	92865.0	3.262	3.59	1.6
Ir	7.2547	11596.5	0.0040	90000.0	3.247	5.27	2.0

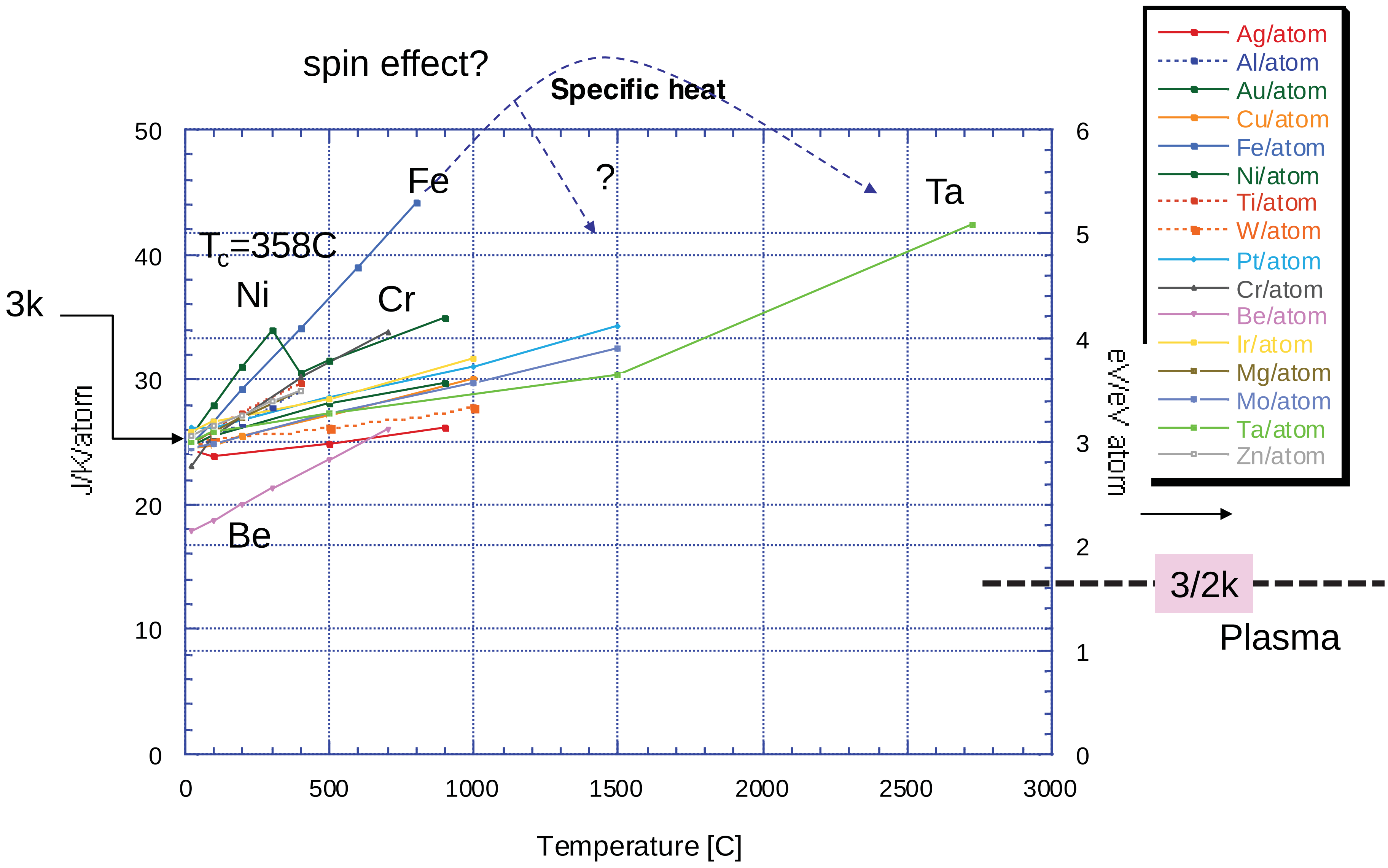
This type of model is useful with real experimental data

$A' = A\exp(-\alpha R_1)$ is the value of V_{sr} at the nearest-neighbour distance;

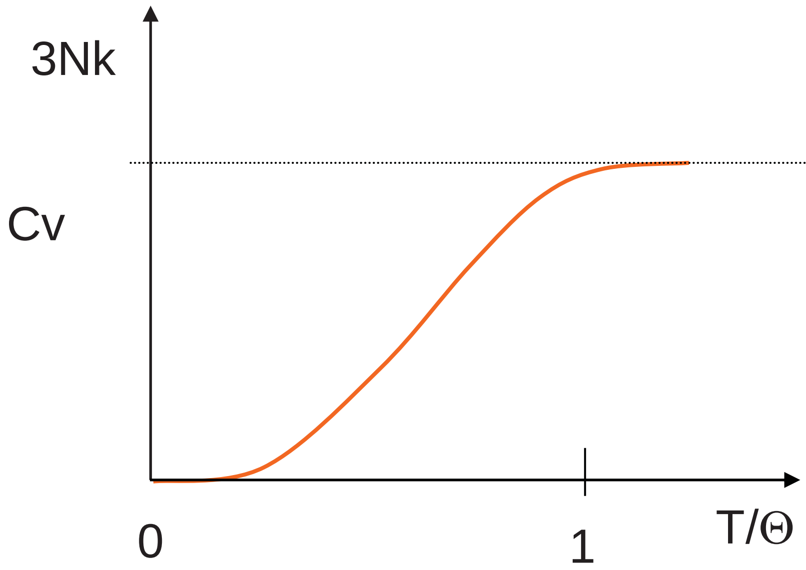
How about specific heat ?

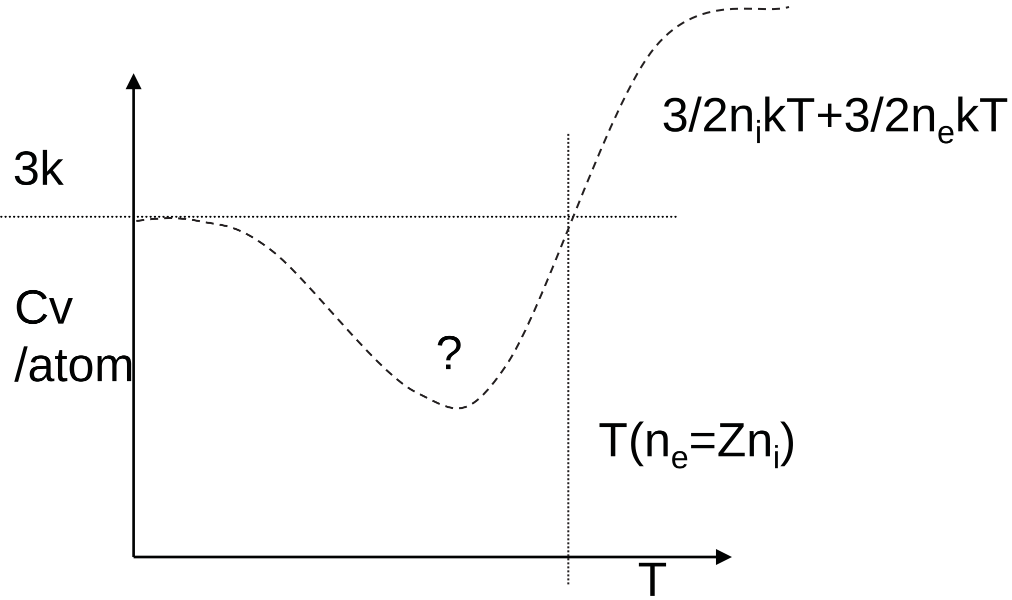


More details in specific heat feature in condensed matter

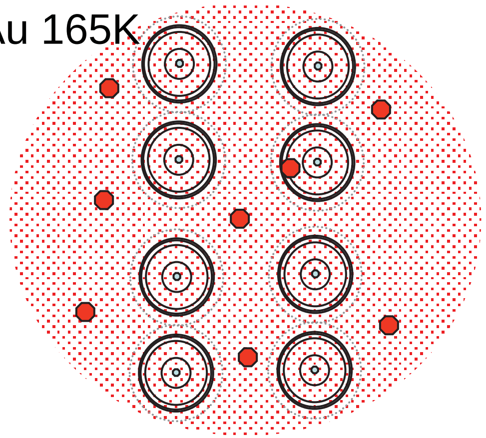


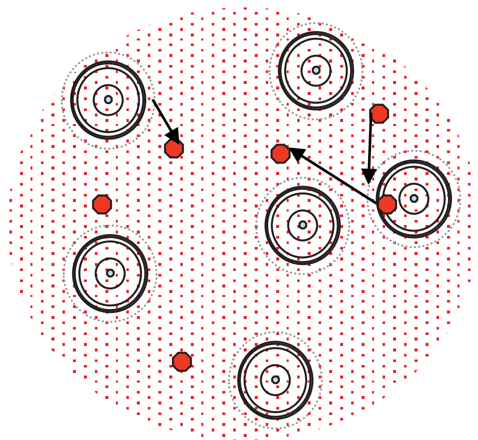
Specific heat in condensed matter and plasma





C 2230K
Be 1440K
Si 645, Cr 630K
Fe 470, Al 428, Ni 343, Ti 420, W 400K
Pt 240, Au 165K





<u>Ordinary plasmas</u>

Melting

Lindemann Law

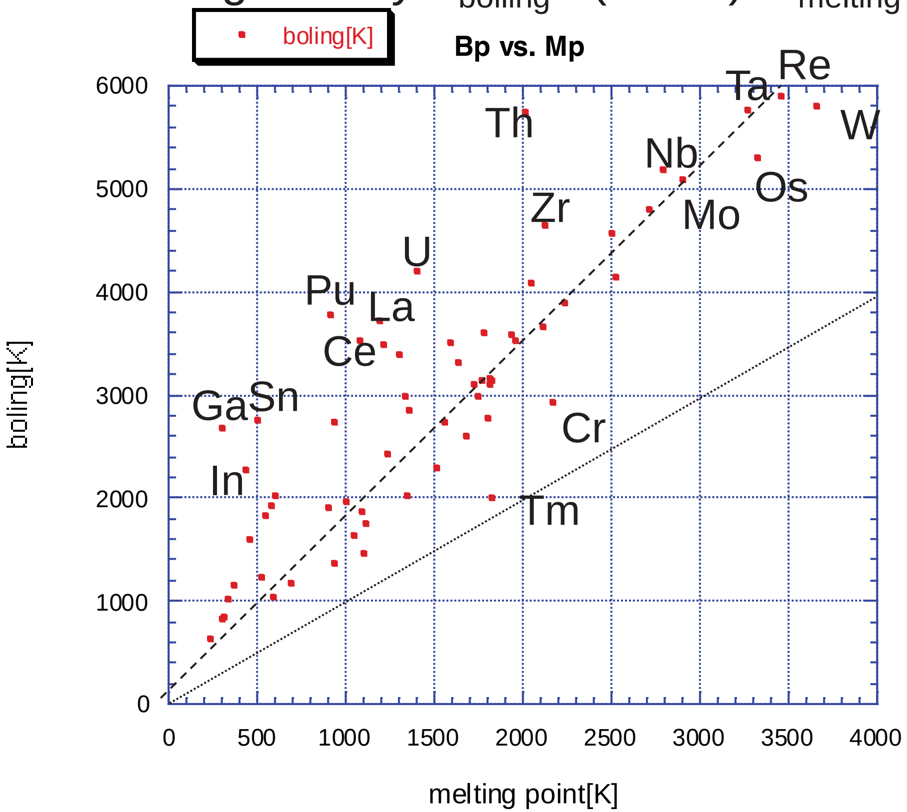
$$T_{melting} = \frac{x_m}{9\hbar^2} Mk\Theta^2 r_s^2 = \frac{\alpha\Theta_{Debye}^2}{\rho^{2/3}}$$

$$\alpha \cong 0.5 A^{5/3}$$

$$<\delta r^2> \sim (r_s/10)^2$$

Boiling

generally $T_{boiling}$ ~(1.7~2)x$T_{melting}$



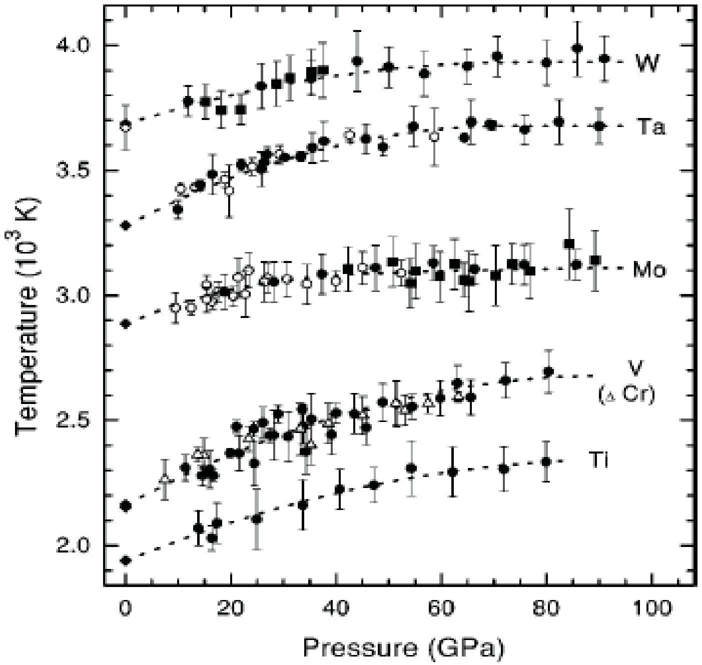
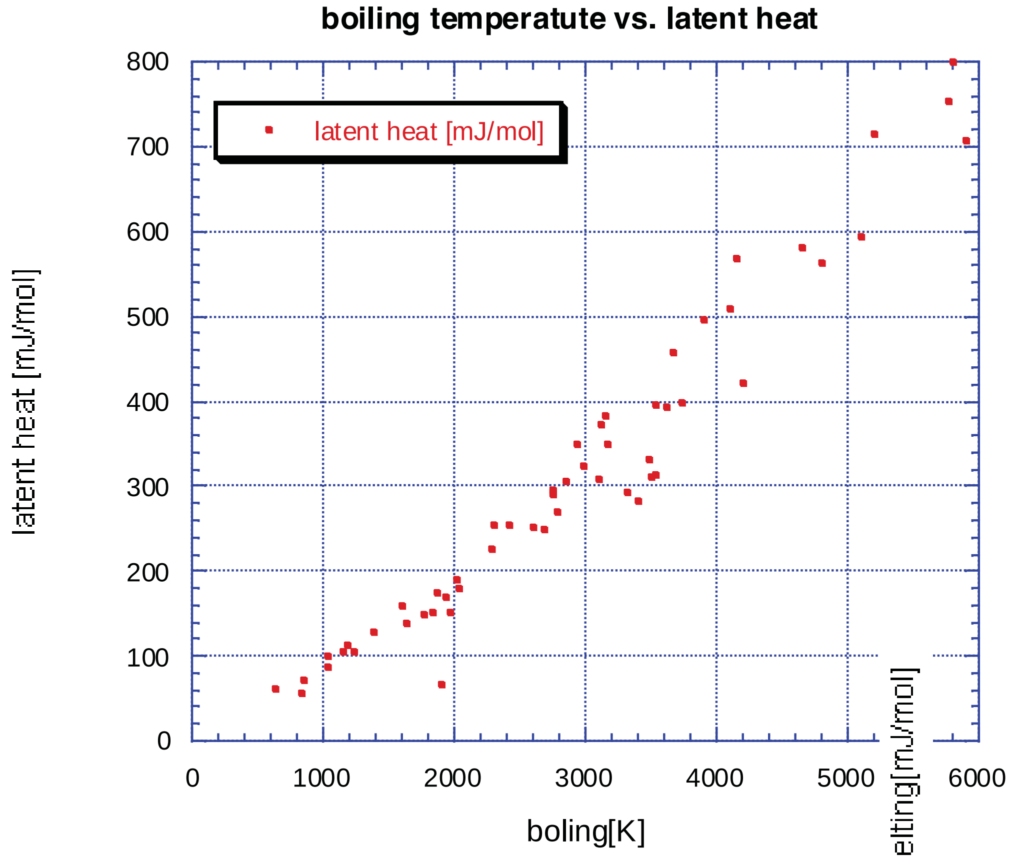
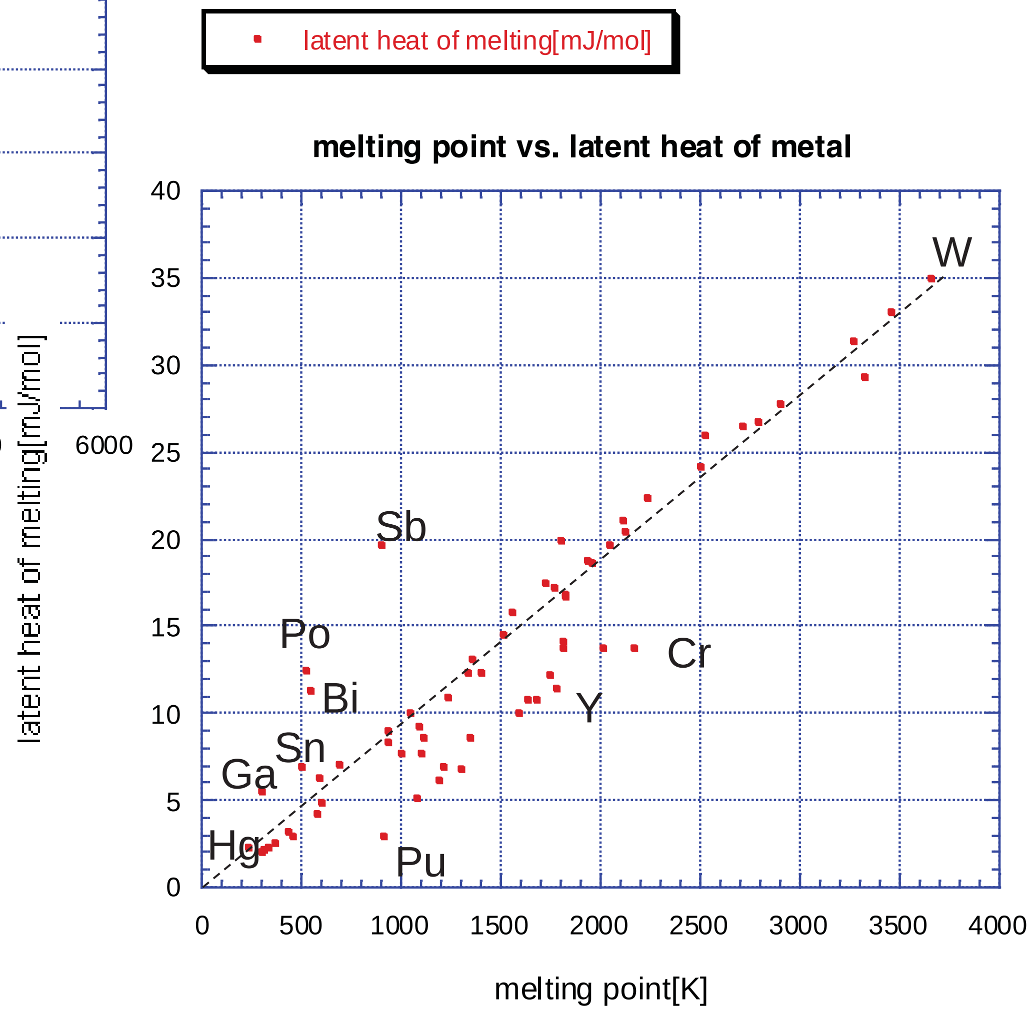


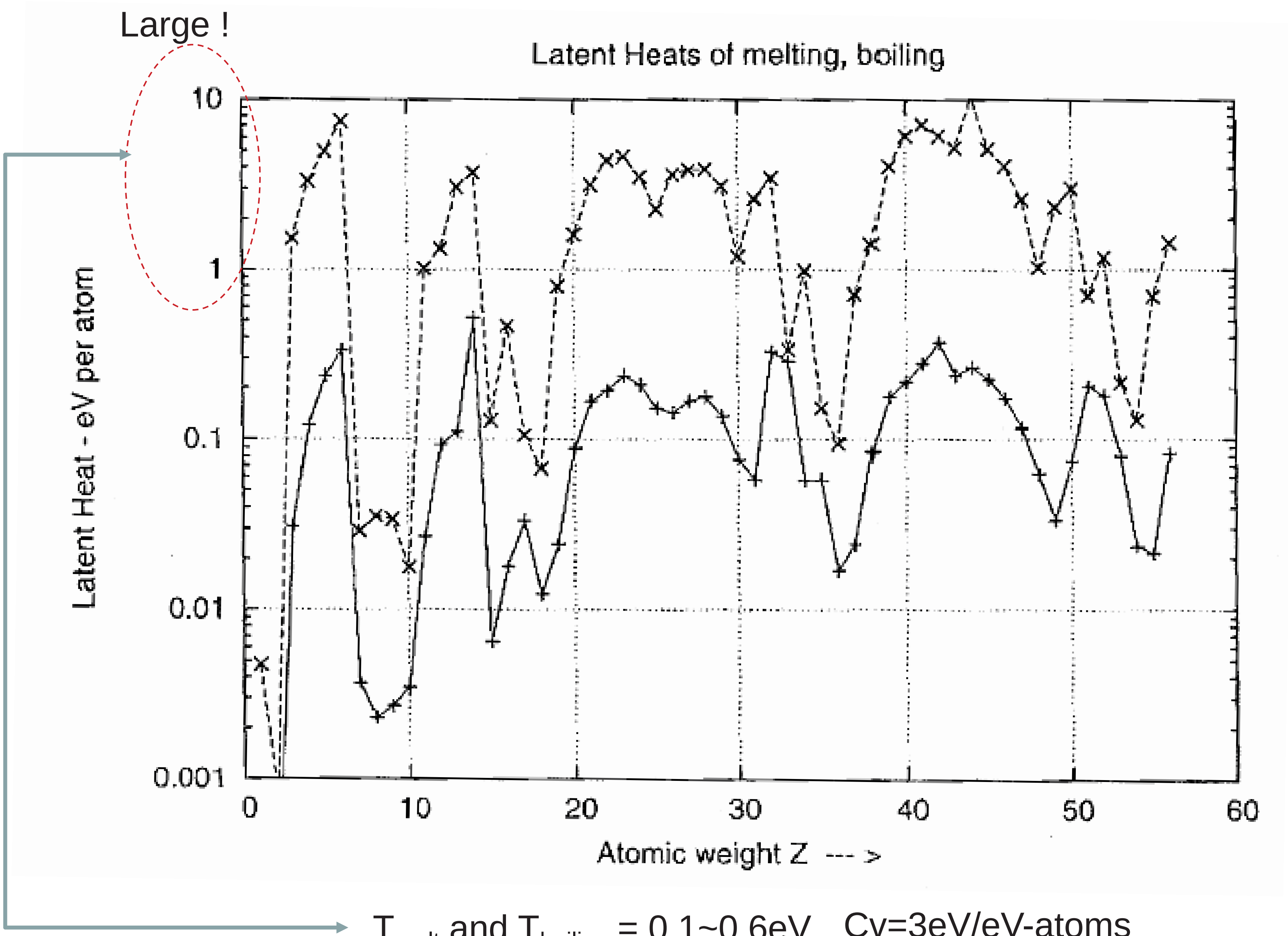
FIG. 2. Melting curves of Mo, Ta, W, V, Ti, and Cr. Solid circles and squares correspond to *in situ* measurements (laser speckle method) in an Ar or Al_2O_3 pressure medium, respectively. Empty circles correspond to the formation of beads as shown in Fig. 1. Open triangles represent the Cr data obtained in situ under Ar. Solid diamonds are 1 atm data. The open diamond represents melting measurements of W in vacuum using identical optics as in the high pressure experiments.

Daniel Errandonea,
PHYSICAL REVIEW B, VOLUME 63, 132104



Simple trend





T_{melt} and $T_{boiling}$ = 0.1~0.6eV   Cv=3eV/eV-atoms

Can we detect m.p. and b.p.?

ns laser heating results

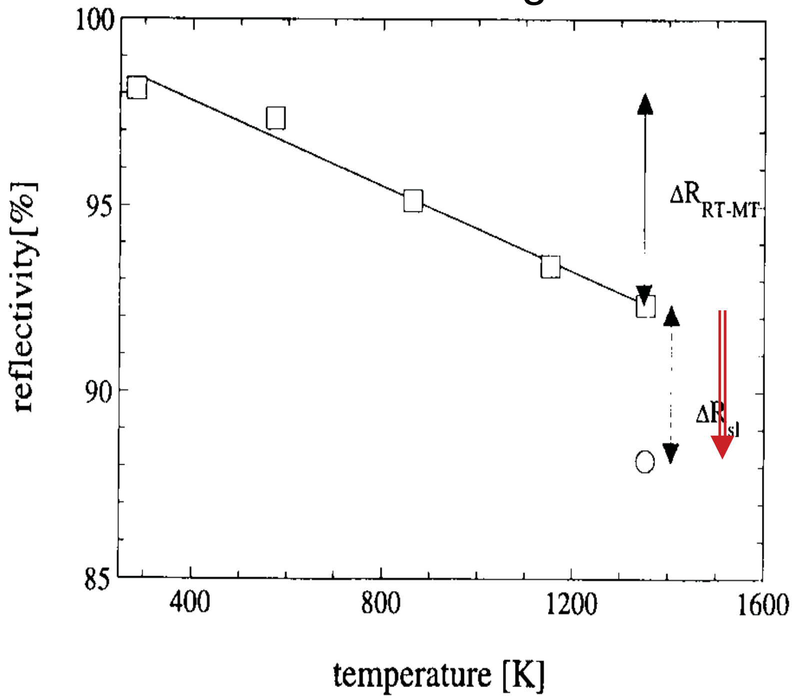
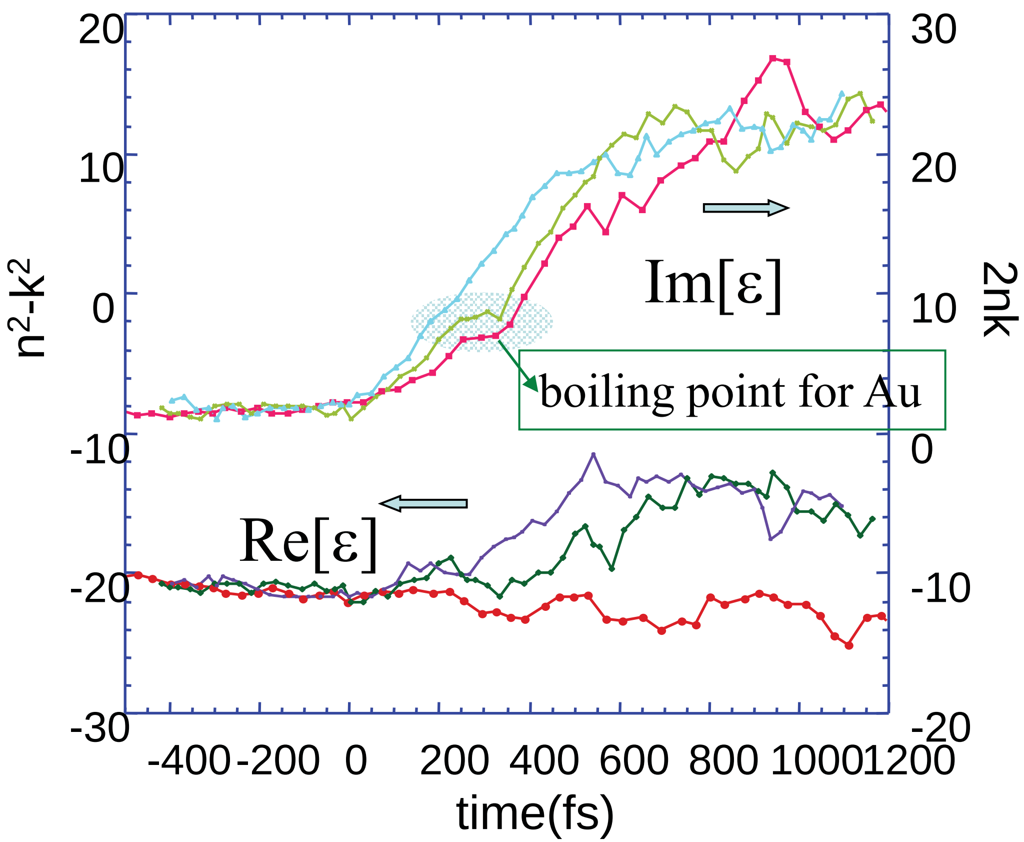


Fig. 2. Calculated reflectivity behaviour of a Cu thin film using the data of Otter [8].

During USP laser heating



*J.Boneberg, et al., Opt. Comm. 174 p/14-149 (2000)

T is Monotonically increase in time.
(during pulse laser duration)